

# On the Concept of Potential in Quantum Field Theory\*

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The literature contains two forms of the two-nucleon potential computed to fourth order in the coupling constant from the gradient coupling of the  $\pi$ -meson to the nucleon field. These are commonly referred to as the Taketani-Machida-Onuma (T.M.O.) and the Brueckner-Watson (B.W.) potentials. The merits of the controversy surrounding this schism are reexamined from first principles starting from the covariant equation for two-nucleons, and it is concluded that the conditions for the applicability of the method leading to the T.M.O. potential are never satisfied in practice. On the other hand, the B.W. potential, suitably altered following recent suggestions by Miyazawa and the author may well yield a suitable approximation in the low energy region.

## § I. Introduction

Recently the author<sup>1)</sup> and Miyazawa<sup>2)</sup> have suggested a new approach to the problem of deducing from meson theory the interaction between two nucleons. In particular they have demonstrated how within a framework, rigorous except for the neglect of a possible meson-meson coupling, the asymptotic form of that interaction for large internucleon separations—usually termed the potential—may be tied completely and directly to parameters deduced from pion-nucleon scattering.

In view of the immodest strength of the above assertion, it may well appear contradictory to have to admit that the functions proposed as “potential” by the two sets of authors differ in an important way. Moreover the nature of that disagreement, usually referred to as the difference between the T.M.O.<sup>3)</sup> and the B.W.<sup>4)</sup> potential, represents the essence of the dispute of longest standing among the *cognescenti* of this subject. The main objective of this note is to attempt to shed definitive light on this controversy. It was felt that the most satisfactory way to proceed would be to reformulate fundamental notions in the soundest manner available, and thus to be able to examine critically alternative and prior attempts. Many of our arguments have been given previously, though we can claim novelty for several observations and for the attempted coherence of the account.

We take as starting point the assertion that the most satisfactory method that we know of in principle of studying the properties of a two-nucleon system is to deduce them from the solutions of the covariant equation for two-fermions<sup>5)</sup>

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$$(G_1 G_2)^{-1} \mathcal{T}(x_1 x_2) = \int I(x_1 x_2; x'_1 x'_2) \mathcal{T}(x'_1 x'_2) d^4 x'_1 d^4 x'_2. \quad (1)$$

Here  $\mathcal{T}(x_1 x_2)$  is the sixteen component amplitude dependent on a pair of space-time coordinates. Since there is a spinor index for every space-time coordinate, the former will be suppressed throughout.  $G_i(x_i, x'_i)$ ,  $i=1, 2$ , are the Feynman or causal propagators for a physical nucleon and thus comprehend fully the self-field of each particle. The exact field theoretical definition of the quantities in eq. (1) as well as a convenient derivation of the exact form of the interaction  $I$  will be described in a succeeding paper. If we restrict ourselves to interactions via the pion field above,  $I$  is nevertheless represented by an infinite series which is a dual expansion both in the number of mesons exchanged and in the non-linear behavior of the mesons. In the applications so far contemplated, the non-linear aspect is neglected. On the other hand, the expansion of  $I$  contains in maximally summed form those self interactions which can be ascribed to an individual nucleon.

The outstanding advantage of eq. (1) are that it can be renormalized and that the necessary choice of an approximate form of  $I$  can be made so as not to destroy either the covariance or the renormalizability. It has been argued against this equation that the amplitude  $\mathcal{T}(x_1 x_2)$  does not admit of any simple statistical interpretation and that it does not lead to a standard boundary value problem, but neither objection can be maintained on principle. Against the former one, it is only necessary to exhibit formulae by which all observables can be related to the amplitude  $\mathcal{T}$ . This has been done by several authors.<sup>(4)</sup> Against the latter objection, it need only be shown that the boundary value problem is well-defined, for example that a suitable normalization condition exists for the discernment of members of the discrete spectrum and that scattering solutions exist. Recent work<sup>(6,7,8)</sup> indicates that this is probably the case.

Indeed the most serious objection against eq. (1) is our impotence to produce solutions for physically applicable situations. In view of this, various efforts have been made to replace eq. (1) by a more Schrödinger-like equation, containing in principle still complete information on the system in question, but one in which consequent approximations would result in a problem for which physically useful solutions could be obtained. To characterize these efforts, let us introduce total and relative coordinates

$$X = \frac{1}{2}(x_1 + x_2), \quad x = x_1 - x_2, \quad (2)$$

and further suppose that we are dealing with an eigenfunction of total energy-momentum vector  $P_u = (\mathbf{P}, W)$ , in particular that  $\mathbf{P} = 0$ . If we then define  $\psi(\mathbf{x}, x_0)$  and  $\varphi(\mathbf{x})$ , respectively, by the equations

$$\mathcal{T}(x_1 x_2) = e^{-iW X_0} \psi(\mathbf{x}, x_0), \quad (3)$$

$$\varphi(\mathbf{x}) = \psi(\mathbf{x}, 0), \quad (4)$$

we first ask whether eq. (1) can be replaced by an equivalent equation of the form

$$[W - H_1(\mathbf{p}) - H_2(-\mathbf{p})] \chi(\mathbf{x}) = \int \mathcal{D}(\mathbf{x}, \mathbf{x}') \chi(\mathbf{x}') d^3 \mathbf{x}' \quad (5)$$



where  $\chi(\mathbf{x})$  either equals  $\varphi(\mathbf{x})$  or is related to it in some known manner,

$$H_i(\mathbf{p}) = [-i\alpha \cdot \mathbf{p} + \beta M]^{(4)} \quad (6)$$

is the free-particle Dirac operator for the  $i^{\text{th}}$  nucleon, and  $\mathcal{G}(\mathbf{x}, \mathbf{x}')$  is the full three-dimensional kernel. Equivalently if we think of  $\chi(\mathbf{x})$ , or rather of its Fourier transform, as decomposed into its constituent positive and negative energy free particle projections and all negative energy components somehow eliminated, we may consider in place of eq. (5), an expression of the form

$$\begin{aligned} [W - E_1(p) - E_2(-p)]\chi(\mathbf{x}) &= (W - 2E(p))\chi(\mathbf{x}) \\ &= \int U(\mathbf{x}, \mathbf{x}') \chi(\mathbf{x}') d^3\mathbf{x}', \end{aligned} \quad (7)$$

where

$$E(p) = (p^2 + M^2)^{1/2} = (-\mathbf{p}^2 + M^2)^{1/2}, \quad (8)$$

and it is henceforth understood that by  $\chi(\mathbf{x})$  we mean a wave-function with positive energy components only.

Many authors<sup>9)</sup> have emphasized that a transition from eq. (1) to eq. (7) is always possible, where, moreover,  $U(\mathbf{x}, \mathbf{x}')$  may be chosen to be independent of the total energy of the system. It is essential here to resume the common thread of the argument.

## § II. Method based on scattering matrix

Let the  $S$  matrix to which eq. (1) gives rise for the scattering of two nucleons from four-momenta  $p_1, p_2$  (and suitable other coordinates which are suppressed) to momenta  $p_1', p_2'$  be represented in the form

$$\begin{aligned} (p_1' p_2' | S | p_1 p_2) &= \delta(\mathbf{p}_1' - \mathbf{p}_1) \delta(\mathbf{p}_2' - \mathbf{p}_2) \\ &\quad - 2\pi i \delta(P_0' - P_0) \delta(\mathbf{P}' - \mathbf{P}) T(\mathbf{p}', \mathbf{p}; \mathbf{P}, P_0), \end{aligned} \quad (9)$$

where we have introduced the total and relative variables

$$\begin{aligned} p_1 &= \frac{1}{2}P + p, & p_1' &= \frac{1}{2}P' + p', \\ p_2 &= \frac{1}{2}P - p, & p_2' &= \frac{1}{2}P' - p'. \end{aligned} \quad (10)$$

In the center of mass frame,  $\mathbf{P} = \mathbf{P}' = 0$ , we have

$$P_0 = 2E(p) = 2E(p'). \quad (11)$$

We may therefore introduce a reduced  $T$  matrix  $t(\mathbf{p}', \mathbf{p})$  defined by

$$T(\mathbf{p}', \mathbf{p}; \mathbf{P} = 0, P_0) = t(\mathbf{p}', \mathbf{p}), \quad (12)$$

regarding the  $P_0$  dependence as indistinct, according to (11).

With

$$u(\mathbf{p}', \mathbf{p}) = (2\pi)^{-3} \int \exp(-i\mathbf{p}' \cdot \mathbf{x}' + i\mathbf{p} \cdot \mathbf{x}) U(\mathbf{x}, \mathbf{x}') d^3\mathbf{x} d^3\mathbf{x}', \quad (13)$$

the corresponding  $T$  matrix associated with the solution of eq. (7) for a scattering situation is the solution of the well-known Lippman-Schwinger integral equation

$$u(\mathbf{p}', \mathbf{p}) = t(\mathbf{p}', \mathbf{p}) - \int \frac{u(\mathbf{p}', \mathbf{p}'') t(\mathbf{p}'', \mathbf{p}) d^3 \mathbf{p}''}{2E(\mathbf{p}) + i\epsilon - 2E(\mathbf{p}'')}. \quad (14)$$

If we now take  $t(\mathbf{p}', \mathbf{p})$  as given from the covariant eq. (1) by eqs. (9) and (12) and look upon eq. (14) as an integral equation for  $u(\mathbf{p}', \mathbf{p})$  then for given  $t(\mathbf{p}', \mathbf{p})$  that equation presumably provides a unique determination of the interaction kernel. An ambiguity which may be serious for bound state problems arises from the fact that eq. (9) determines  $T$  only up to expressions which vanish on the energy shell, which has a similar consequence for  $u(\mathbf{p}', \mathbf{p})$ . We may at least assert, however, that there exist a class of interaction functions, non-local but energy independent, which are equivalent with respect to scattering consequences. In our future discussion, we shall suppose a definite form of  $u$  derived from a definite choice of  $T$ . In practice such a choice usually presents itself in a natural way as the one which emerges directly from the calculation that leads to (9) without any reexpression by the gratuitous use of the energy conservation condition.<sup>10)</sup>

In practice again  $t(\mathbf{p}', \mathbf{p})$  is given as some form of power series simply related to the kernel  $I$  of eq. (1). Equation (14) is itself then solved by iteration methods giving rise to a new power series whose convergence properties are a matter of fundamental interest. Though we do not wish to enter here into the further questions, it should also be pointed out that other observables of the theory, for example, electromagnetic moments, are represented by similar power series.

### § III. Method based on approximate relative time dependence

Some light is shed on questions of convergence by the discussion of an alternative mode of obtaining a three-dimensional equation. Its end result replaces eq. (7) by the form

$$(W - 2E(\mathbf{p})) \varphi(\mathbf{x}) = \int V(\mathbf{x}, \mathbf{x}'; W) \varphi(\mathbf{x}') d^3 \mathbf{x}', \quad (15)$$

in which the kernel  $V(\mathbf{x}, \mathbf{x}'; W)$  is in general a transcendental function of the total energy  $W$  and again a power series of some kind. The basic assumption of this method is a separability hypothesis, that the relative time amplitude  $\psi(\mathbf{x}, x_0)$  may be expressed in the form

$$\psi(\mathbf{x}, x_0) = \int J(\mathbf{x}, \mathbf{x}'; x_0; W) \varphi(\mathbf{x}') d^3 \mathbf{x}'. \quad (16)$$

By employing eq. (16) on the right hand side of eq. (1), the latter itself may be used in an iterative procedure to determine  $J$  once the zeroth approximation has been stated. The starting point is taken to be the relative time dependence characteristic of a situation in which the interaction  $I$  is itself instantaneous, in which case the separability hypothesis (16) is known to be fulfilled exactly.<sup>12)</sup>



We shall omit further details since they have been recounted exhaustively in the literature.<sup>11)</sup> The resulting equation (15) has however a pair of properties which are essential to the present discussion. For the first of these we consider the Fourier transform of  $V(x, x'; W)$  which we denote by  $v(\mathbf{p}', \mathbf{p}; W)$ . The associated transition amplitude  $t(\mathbf{p}', \mathbf{p}; W)$ , as determined from the analogue of (14),

$$v(\mathbf{p}', \mathbf{p}; W) = t(\mathbf{p}', \mathbf{p}; W) - \int \frac{v(\mathbf{p}', \mathbf{p}''; W) t(\mathbf{p}'', \mathbf{p}; W) d^3 \mathbf{p}''}{W + i\epsilon - 2E(\mathbf{p}'')}, \quad (17)$$

agrees for  $W = 2E(p) = 2E(p')$  with  $t(\mathbf{p}', \mathbf{p})$  of eq. (12). From this point of view eq. (15) is fully as satisfactory as eq. (7).

The second point concerns the relationship between eq. (7) and eq. (15).<sup>13)</sup> We here describe a procedure by which the transition between the two can be effected. We ask that eq. (15) be equivalent (in momentum space) to an equation of the form

$$(W - 2E(p)) \chi(\mathbf{p}) = \int v_e(\mathbf{p}, \mathbf{p}') \chi(\mathbf{p}') d^3 \mathbf{p}', \quad (18)$$

where  $\chi(\mathbf{p})$  is taken to be a normalized amplitude. We are also free to require for the amplitude of (15)

$$\int \varphi^*(\mathbf{p}) \varphi(\mathbf{p}) d^3 \mathbf{p} = 1, \quad (19)$$

though this necessarily implies a non-unit norm for the state vector from which  $\varphi$  was derived. Under these circumstances we can seek a unitary connection

$$\begin{aligned} \varphi(\mathbf{p}) &= \int \Lambda(\mathbf{p}, \mathbf{p}'; W) \chi(\mathbf{p}') d^3 \mathbf{p}' \\ \Lambda^\dagger(W) \Lambda(W) &= 1. \end{aligned} \quad (20)$$

$\Lambda(W)$  and  $v_e$  are then determined by the matrix equation

$$\begin{aligned} \langle \mathbf{p} | \{ \Lambda^\dagger(W) v(W) \Lambda(W) + \Lambda^\dagger(W) 2E \Lambda(W) \} | \mathbf{p}' \rangle \\ - 2E(p) \delta(\mathbf{p} - \mathbf{p}') = v_e(\mathbf{p}, \mathbf{p}'), \end{aligned} \quad (21)$$

and by (20). To eliminate the explicit energy dependence of the left hand side of (21) we carry out the power series developments

$$\Lambda^\dagger(W) = \Lambda^\dagger(2E(p)) + (W - 2E(p)) \frac{\partial \Lambda^\dagger}{\partial W} \Big|_{W=2E(p)} + \dots, \quad (22)$$

$$\Lambda(W) = \Lambda(2E(p')) + (W - 2E(p')) \frac{\partial \Lambda}{\partial W} \Big|_{W=2E(p')} + \dots, \quad (23)$$

$$v(W) = v(E(p) + E(p')) + (W - E(p) - E(p')) \frac{\partial v}{\partial W} \Big|_{W=E(p)+E(p')} + \dots. \quad (24)$$

Inserting (22)–(24) into (21), and noting that the expectation value of (21) in the state  $\chi(\mathbf{p})$  is implied, we may replace  $W - 2E(p)$  on the left and  $W - 2E(p')$  on

the right by  $v_e$ . We obtain an equation for the determination  $v_e(\mathbf{p}, \mathbf{p}')$  and the matrix elements of  $A$  and its energy derivatives to be solved in conjunction with (20). In practice the power series method in the coupling constant or some small modification thereof is the only one available. For example, an equation which is correct to fourth order in the coupling constant (ignoring the terms involving  $E(p)$ ) but which also implies selected higher order effects is given by the expression

$$v_e(\mathbf{p}, \mathbf{p}') = v(\mathbf{p}, \mathbf{p}'; E(\mathbf{p}) + E(\mathbf{p}')) + \langle \mathbf{p} | \frac{1}{2} \left\{ v_e, \frac{\partial v(W)}{\partial W} \right\}_{E(\mathbf{p}) + E(\mathbf{p}')} | \mathbf{p}' \rangle. \quad (25)$$

An equation such as (25) will be compared below with eq. (24) which suggests itself as the expansion to be used directly in (15).

To pursue the discussion further at this stage, we must ask for the connection between  $v_e(\mathbf{p}, \mathbf{p}')$  and the kernel  $u(\mathbf{p}, \mathbf{p}')$  associated with eq. (7). Our general discussion should make it clear that there is no *a priori* reason that they be identical term by term. What is evident is that they again lead to the same scattering matrix on the energy shell. For the same reason that this is true they must also yield the same form of the adiabatic potential, to the discussion of which we must now turn.

#### § IV. Comparison of methods and conclusions

To our knowledge, no fruitful solutions of eqs. (7) or (15) have been obtained for a non-local kernel. Though this reality must eventually be faced, efforts to date have been directed toward replacing  $U(\mathbf{x}, \mathbf{x}')$ , for example, by a static potential,

$$U(\mathbf{x}, \mathbf{x}') \rightarrow U(\mathbf{x}) \delta(\mathbf{x} - \mathbf{x}') \quad (26)$$

a replacement that is justifiable asymptotically and combining this with a phenomenological treatment of the interaction at small distances. In momentum space this corresponds to the reduction of  $u(\mathbf{p}', \mathbf{p})$  to a function  $v(\mathbf{p}' - \mathbf{p})$  of the momentum transfer alone. This can be brought about and is justified whenever both  $\mathbf{p}$  and  $\mathbf{p}'$  can be considered small compared to  $M$  and thus differences such as  $E(p) - E(p')$  can be neglected.<sup>14)</sup> Since this same neglect is possible on the energy shell, where  $u(\mathbf{p}', \mathbf{p})$  and  $v_e(\mathbf{p}', \mathbf{p})$  coincide, it follows that these kernels also give rise to the same adiabatic potential. If we further restrict ourselves to the coupling of  $p$ -wave mesons (gradient coupling) with nucleons, work only up to the exchange of two mesons, ignoring all radiative corrections other than those which produce the renormalized coupling constant from the bare one, the resulting local interaction constitutes what has been termed the T. M. O. potential.

When we turn next to the corresponding specialization of the kernel  $V(\mathbf{x}, \mathbf{x}'; W)$  of eq. (15), in addition to the requirement stated in the previous paragraph, necessary for the passage to the adiabatic-limit, two additional conditions must be met. These are that we may ignore the differences  $W - 2M$  and  $E(p) + E(p') - 2M$ , again compared to a typical meson energy. With these neglects, we find as before

$$V(\mathbf{x}, \mathbf{x}'; W) \rightarrow V(\mathbf{x}) \delta(\mathbf{x} - \mathbf{x}'). \quad (27)$$



Under the same coupling conditions as stated in the previous paragraph, the resulting local interaction is what has been termed the B. W. potential.

We now compare the two potentials. We shall be forced to conclude that under conditions for which the adiabatic approximation is valid, if the T. M. O. and B. W. potentials differ in their predictions of the interaction in any state of the two-nucleon system, then if either is correct, it must be the B. W. potential. This conclusion follows from the considerations associated with eqs. (18) — (25). Thus the expansion (24) is justified only if the "adiabatic" hypothesis is warranted for  $V(\mathbf{x}, \mathbf{x}'; W)$ . This may well not be the case for energetic collisions of two nucleons, as an example for 100 Mev available energy in the center of mass system. As a prototype of what occurs at low energies, however, Brueckner and Watson<sup>4)</sup> have estimated the relative size of the second term of eq. (24) compared to the first for the Deuteron problem and found it to be quite negligible. However, when treated as in the transition to eq. (25), where it gives rise in a perturbation treatment to a contribution to the fourth order potential (exchange of two mesons), it is well known to yield a large, indeed dominant contribution to the central potential in the triplet even state. In this transition, however, the second term of (24) is replaced by a power series in the coupling constant of which the fourth order term is only the first one. The series in question must be either poorly or non-convergent in the region of interest. This is illustrated for eq. (25) in the Appendix. We further conclude that the method of treating the two-nucleon problem based on an energy-independent kernel,  $V(\mathbf{x}, \mathbf{x}')$  where the latter is understood as a power series is not applicable to pseudoscalar mesons in any practical sense.

To complete the discussion we must endeavor to dispose of the main argument remaining which is adduced in the attempt to refute the above conclusions.<sup>10)</sup> It is based on the historically oft-used adiabatic method for the treatment of the two nucleon problem.<sup>16)</sup> In this method one computes the interaction energy of two nucleons at rest and uses this as the potential energy in a non-relativistic Schrödinger equation. Now if one applies this to the neutral scalar meson theory, it is well-known that the second order Yukawa potential indeed provides the exact static interaction energy. If one now turns to the methods of this paper it is found that  $U(\mathbf{x}, \mathbf{x}')$  of eq. (7) does indeed have this value in the adiabatic limit, whereas  $V(\mathbf{x}, \mathbf{x}'; W)$  of eq. (15) gives rise to finite corrections of fourth and higher order. From this it is concluded that the adiabatic limit has not been properly taken in the latter case.

We now insist that the argument given above *begs the question*. For it employs as a club the adiabatic method, whereas it is precisely the conventional application of the latter which is in contention. It should be evident by now that the question just raised can be settled only by appeal to non-adiabatic methods such as we have exposed in this paper. Alternatively phrased, there is no *a priori* reason that the exact answer of the adiabatic method yield the most reliable simple approximation to the fully relativistic situation. Indeed, we believe that we have established that for pseudoscalar meson theory, this is definitely not the case.

In the work of Miyazawa<sup>1)</sup> and the author<sup>2)</sup> alluded to in the introduction, all

results were obtained through the intermediary of an  $S$ -matrix formalism similar to that which leads to eq. (7). However, the work of the author was modified so as to take account of what was asserted to be the known form of eq. (15) as applied to the formalism of that paper. In the paper which follows, we verify those assertions, at the same time extending the previous formalism.

Our general conclusion is therefore that eq. (7), with  $U(\mathbf{x}, \mathbf{x}')$  represented by a power series in the number of mesons exchanged is *never* applicable. We have, of course, not thereby established the validity of eq. (15). Arguments given elsewhere,<sup>1)</sup> however, lead us to believe that with  $V(\mathbf{x}, \mathbf{x}'; W)$  determined by the method of the following paper, it should have a useful domain of application and that its "adiabatic form" may be valid at low energies.

Of course, the use of (15) with its energy dependent interaction implies that the formula for other observables must be suitably modified, and the utility of the complete program based on (15) becomes tied to a rapid convergence of the series representing these quantities as well as that which describes the interaction. This latter question will be dealt with on a future occasion.

The author takes this opportunity to recall with pleasure stimulating conversations on the topic of this note with Drs. K. Brueckner, D. Feldman, N. Fukuda, S. Machida, and M. Ruderman.

## Appendix

We shall illustrate here how the procedure leading from (24) to (25) of the text gives rise to results which gainsay the fundamental assumptions on which it is based. Working only to the second order in the coupling constant for  $v(\mathbf{p}, \mathbf{p}'; W)$  and passing to the adiabatic limit, we easily obtain from (25) the following equation for the determination of  $v_e(\mathbf{r})$ <sup>13)</sup>,

$$v_e(\mathbf{r}) = v_2(\mathbf{r}) - v_2'(\mathbf{r}) v_e(\mathbf{r}) \quad (\text{A} \cdot 1)$$

where

$$v_2(\mathbf{r}) = \frac{1}{3} (f^2/4\pi) \mu \boldsymbol{\tau}_1 \cdot \boldsymbol{\tau}_2 (e^{-x}/x) \times [\sigma_1 \cdot \sigma_2 + x^{-2} (x^2 + 3x + 3) S_{12}], \quad (\text{A} \cdot 2)$$

$$v_2'(\mathbf{r}) = \frac{1}{3} (f^2/4\pi) \boldsymbol{\tau}_1 \cdot \boldsymbol{\tau}_2 (2/\pi) \{ \sigma_1 \cdot \sigma_2 [ (K_1(x)/x) - K_0(x) ] - S_{12} [ (2K_1(x)/x) + K_0(x) ] \}. \quad (\text{A} \cdot 3)$$

We have chosen units in which  $\hbar=c=1$ ,  $S_{12}$  is the tensor operator,  $K_1(x)$  and  $K_0(x)$  are the Hankel functions of imaginary argument and  $x=\mu r$  is the separation measured in units of the pion Compton wavelength.

We shall solve (A·1) in the given approximation for the triplet even central force. We write in this state

$$v_e(\mathbf{r}) = G_1(r) + G_2(r) S_{12} \quad (\text{A} \cdot 4)$$

and further define



$$\begin{aligned}
A_1 &= -(f^2/4\pi) \mu(e^{-x}/x), \\
A_2 &= (f^2/4\pi) \mu(e^{-x}/x^3) (x^2 + 3x + 3) \\
\alpha_1 &= (f^2/4\pi) (2/\pi) [(K_1(x)/x) - K_0(x)] \\
\alpha_2 &= -(f^2/4\pi) (2/\pi) [(2K_1(x)/x) + K_0(x)].
\end{aligned} \tag{A.5}$$

By means of the relation

$$\begin{aligned}
S_{12}^2 &= 6 + 2\sigma_1 \cdot \sigma_2 - 2S_{12} \\
&\rightarrow 8 - 2S_{12}
\end{aligned} \tag{A.6}$$

for the triplet spin state, we easily obtain two linear equations for  $G_1$  and  $G_2$ , with solution

$$G_1(r) = \frac{A_1(1 - \alpha_1 + 2\alpha_2) - 8\alpha_2 A_2}{(1 - \alpha_1)(1 - \alpha_1 + 2\alpha_2) - 8\alpha_2^2}, \tag{A.7}$$

$$G_2(r) = \frac{-A_2 + \alpha_2 G_1}{1 - \alpha_1 + 2\alpha_2}. \tag{A.8}$$

For a typical value of  $(f^2/4\pi) = .1$ , two observations can be made. Even at the force range  $|G_1(r)| \gg |A_1(r)|$ , the latter measuring the strength of the second order central potential. Moreover  $G_1(r)$  itself blows up at  $x \sim .7$ . The comparison of  $G_2(r)$  with  $A_2(r)$  though not quite as catastrophic, is qualitatively similar with respect to the second observation. In any event this is sorry behavior for the quantity  $|G_1(r) - A_1(r)|$  which should be small compared to  $|A_1(r)|$  itself and which apparently<sup>4)</sup> has this property when treated in the form (24), the approximate passage to (25) being avoided completely because of its demonstrated invalidity.

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## A Comparison of Plane Wave Solutions in General Relativity with Those in Non-Symmetric Theory

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Exact plane wave solutions of the field equations in general relativity and those of the field equations in non-symmetric unified field theory are obtained under similar conditions for the case in which electromagnetic fields are present. Main properties of both solutions are compared with each other. The most important result is that, as far as the solutions dealt with in this paper are concerned, the space-time is closely connected with the electromagnetic field in general relativity, while in non-symmetric unified field theory the structure of the Riemannian space-time is determined quite independently of the electromagnetic field.

### § 1. Introductory remarks

The object of this paper is to compare some wave solutions of field equations in the general theory of relativity with those of field equations in the non-symmetric unified field theory of Einstein, and to offer some materials for the solution of the problem which of the two theories is more appropriate for describing natural phenomena. These two theories will hereafter be referred to as GR and NS respectively.

The field equations in NS coincide formally with the purely gravitational equation for empty region in GR when the non-symmetric fields (fundamental tensor and coefficients of connection) are reduced to the symmetric ones, and the difference between both theories appears only when these fields are not symmetric. To express it in terms of physics, this means that the both equations differ only in some region where there exists some other field besides gravitation. Therefore we shall confine ourselves to some region in which both gravitational and electromagnetic fields are present.

It is not easy, however, to compare the solutions of both theories 'completely'. This difficulty comes mainly from the fact that the fundamental ideas with which both theories are constructed are not the same and the characters of both field equations are different from each other. Main differences are as follows: (1) In GR, the source terms for gravitational field are contained in the right-hand side of the field equations, but on the contrary, all such terms are absorbed in the left-hand side of the field equations in NS. (2) In GR, it is generally accepted that some kinds of singularities represent the existence of matter and that solutions containing such singularities are admitted. On the other hand, only those solutions that are regular everywhere are required in NS.

In this paper, therefore, as the first step of the comparison, we shall compare both

solutions 'in the small'. Thus, we restrict the problem in some limited portion of the space-time, and do not deal with the problem concerning the singularities of the solutions, nor the one whether we can extend the solutions in question into the whole space-time.

In making such a comparison it is very desirable to deal with exact solutions. The both field equations, however, are considerably complicated, therefore it is not so easy to obtain their solutions, especially exact ones. Up to the present, many investigations have been done in GR concerning its field equations and various solutions obtained. However, if we exclude those for cosmological problems, there have been obtained few exact non-static solutions. Especially, little is known concerning exact wave solutions for the case in which electromagnetic fields are present. As to NS, the circumstances are more unsatisfactory, and it will not be too much to say that we have no knowledge concerning such a wave solution. This is the present situation of the exact wave solutions.

Recently the present writer has succeeded in obtaining some exact solutions of the both field equations corresponding to the system where matter does not exist at all and the electromagnetic field is composed of plane waves propagating in one direction. The details of the calculation are given in the references.<sup>1,2,3</sup> In what follows, we shall explain these solutions and then compare some main features of the both solutions in conformity with the considerations given above.

## § 2. Field equations

### (a) Field equations in GR

As was stated in § 1, we are going to deal with a system composed of purely radiational fields alone. Accordingly, not only the energy tensor due to mass but also the charge-current vector must vanish everywhere. Thus, the field equations become

$$\begin{cases} G_{ij} = -8\pi E_{ij}, & F^{ij}_{;j} = 0, \\ F_{ij;k} + F_{jk;i} + F_{ki;j} = 0, & (i, j, \dots = 1, \dots, 4), \end{cases} \quad (\text{GRE})$$

where  $G_{ij}$  is the Ricci tensor of the space-time,  $E_{ij}$  is the electromagnetic energy tensor,  $F_{ij}$  is the antisymmetric tensor representing the electromagnetic strength, and a semicolon followed by an index denotes covariant derivative. It is needless to say that the last two equations of (GRE) are the generalized Maxwell equations and are reduced to the usual Maxwell equations in the Minkowski space-time.

### (b) Field equations in NS

The strong field equations in NS are as follows:

$$\begin{cases} g_{ij;\pm} = 0, & \Gamma_{ij}^j = 0, \\ R_{ij} = 0, & (i, j, \dots = 1, \dots, 4) \end{cases} \quad (\text{NSE})$$

where  $g_{ij}$ ,  $\Gamma_{ij}^k$  and  $R_{ij}$  are the non-symmetric fundamental tensor, the coefficients of non-symmetric connection and the generalized Ricci tensor respectively. The notations  $\pm$  and  $-$  under indices indicate the kinds of covariant derivatives.

In (NSE), in view of later convenience, we have used the same notation  $g_{ij}$  to



denote the non-symmetric fundamental tensor as the symmetric fundamental tensor in GR. But there will be no apprehension of confusion.

It should be noted that, in NS, the field equations are given by the above (NSE) irrespective of the kind of system under consideration.

### § 3. Identifications of mathematical expressions with physical quantities

In GR, the physical quantities are well defined and their mathematical expressions well known. The gravitational potential is expressed by the fundamental tensor, and further, as was stated in last section, the electromagnetic strength, the electromagnetic energy and the charge-current vector are expressed respectively by  $F_{ij}$ ,  $E_{ij} = g_{ij} F_{kl} F^{kl} / 4 - F_{ik} F_{jl} g^{kl}$  and  $F^{ij}{}_{;j}$ .

In NS, on the contrary, there is no rule of the correspondence between the mathematical expressions and the physical quantities, which is universally accepted. Without this rule, however, we have no way of continuing our reasoning. Therefore, following the opinions of some authors, e. g. Einstein,<sup>3)</sup> Hlavatý,<sup>4)</sup> Ikeda,<sup>5)</sup> we shall make the assumptions :

(i)  $h_{ij} \equiv g_{ij}$  plays the same role as  $g_{ij}$  in GR. That is,  $h_{ij}$  expresses the gravitational potential.

(ii) The electromagnetic strength  $F_{ij}$  is given by

$$F_{ij} = \sqrt{-g} \epsilon_{ijlm} g^{lm} / 2, \quad (3.1)$$

where  $g = \det g_{ij}$ ,  $g^{ij}$  is the tensor defined from  $g_{ij}$  by  $g_{ij} g^{kj} = \delta_i^k$ , and  $\epsilon_{ijlm}$  is the relative tensor whose components are antisymmetric with respect to every pair of indices and  $\epsilon_{1234} = 1$ .

(iii) The charge-current density vector is expressed by

$$\mathbf{j}^i = \epsilon^{ijkl} g_{jk,l} / 3!, \quad (3.2)$$

where a comma followed by an index denotes usual partial derivative and  $\epsilon^{ijkl}$  is the relative tensor similar to the  $\epsilon_{ijkl}$ .

Here we shall give two remarks concerning these assumptions.

(1) We start with the identification (3.1). However, even if we adopt the relation

$$F_{ij} = g_{ij} \quad (3.3)$$

in place of (3.1), we arrive at the same results so far as the solutions dealt with in this paper are concerned. Of course, we can not expect such a coincidence in more general cases.

(2) Strictly speaking,  $F_{ij}$  defined by (3.1) is not an ordinary tensor but a  $W$ -tensor. In order to make  $F_{ij}$  an ordinary tensor, we have to multiply the right-hand member of (3.1) by a suitable factor. Detailed investigation concerning this fact has

## § 5. Exact solutions

In this section we shall explain exact solutions which are obtained from Assumptions 1 and 2.

### (a) Solution of (GRE)

Under our two assumptions, the general solution of (GRE) is as follows:

(GRS)  $g_{ij}$  is given by (4.1), and  $F_{ij}$  is given by

$$F_{ij} \begin{cases} E: & F_{14}=\sigma, & F_{24}=-\rho, & F_{34}=0, \\ H: & F_{23}=\rho, & F_{31}=\sigma, & F_{12}=0, \end{cases} \quad (5.1)$$

where  $\rho$ ,  $\sigma$  as well as  $A$ ,  $B$ ,  $C$ ,  $D$  are functions of  $Z$ . Six functions are required to satisfy only one condition,

$$P = -8\pi (A\rho^2 + 2D\rho\sigma + B\sigma^2)/m, \quad (5.2)$$

where

$$m = AB - D^2, \quad (5.3)$$

$$P = (Av - 2Dw + Bu)/m$$

$$= \bar{m}/2m - \bar{m}^2/4m^2 - \bar{m}\bar{C}/2mC - (\bar{A}\bar{B} - \bar{D}^2)/2m, \quad (5.4)$$

$$\begin{cases} 2u = \bar{A} - (B\bar{A}^2 + A\bar{D}^2 - 2D\bar{A}\bar{D})/2m - \bar{A}\bar{C}/C, \\ 2v = \bar{B} - (A\bar{B}^2 + B\bar{D}^2 - 2D\bar{B}\bar{D})/2m - \bar{B}\bar{C}/C, \\ 2w = \bar{D} - (B\bar{A}\bar{D} + A\bar{B}\bar{D} - D\bar{A}\bar{B} - D\bar{D}^2)/2m - \bar{C}\bar{D}/C, \end{cases} \quad (5.5)$$

and a bar over a kernel letter means derivative with respect to  $Z$ , e. g.  $\bar{A} = dA/dZ$ ,  $\bar{\bar{A}} = d^2A/dZ^2$ .

We can consider the relation (5.2) as an algebraic equation to determine  $F_i$  (i. e.  $\rho$ ,  $\sigma$ ) when  $g_{ij}$  (i. e.  $A$ ,  $B$ ,  $C$ ,  $D$ ) is given, or alternatively, as a differential equation to determine  $g_{ij}$  when  $F_{ij}$  is given. Anyhow, it is only one condition for six functions and accordingly it admits solutions with great arbitrariness. Let  $(\rho, \sigma)$  be a solution of (5.2). Then, since  $(\rho, \sigma)$  is contained in (5.2) in the combined form

$$\alpha(\rho, \sigma) = A\rho^2 + 2D\rho\sigma + B\sigma^2 \quad (\geq 0), \quad (5.6)$$

there exist other  $(\rho, \sigma)$ 's which satisfy (5.2) together with the same  $g_{ij}$ . Detailed mathematical research concerning such problems is given in the reference.<sup>1)</sup>

### (b) Solution of (NSE)

Using the assumptions stated in the last section and one additional technical assumption which is explained in the Appendix we can solve (NSE). The solution is as follows<sup>2)</sup>:

(NSS)  $h_{ij} \equiv g_{ij}$  and  $f_{ij} \equiv g_{ij}$  are given respectively by (4.1) and

$$f_{13} = -f_{14} = \rho', \quad f_{23} = -f_{24} = \sigma', \quad f_{12} = f_{34} = 0, \quad (5.7)$$

where  $\rho'$  and  $\sigma'$  are arbitrary functions of  $Z$ , and  $A$ ,  $B$ ,  $C$ ,  $D$  are subject to only one condition



$$P=0, \quad (5.8)$$

$P$  being the quantity given by (5.4).  $(\rho', \sigma')$  and  $(A, B, C, D)$  are quite independent of each other.

For this solution,  $F_{ij}$  is of the same form as that in the (GRS), i. e.

$$F_{12}=F_{34}=0, \quad F_{14}=F_{31}=\sigma, \quad -F_{24}=F_{23}=\rho \quad (5.9)$$

where we have put

$$\rho = (B\rho' - D\sigma')/\sqrt{m}, \quad \sigma = (A\sigma' - D\rho')/\sqrt{m}. \quad (5.10)$$

Further, if we calculate the charge-current density vector  $\mathbf{j}^i$  defined by (3.2), we have identically

$$\mathbf{j}^i = 0. \quad (5.11)$$

Here we shall add two remarks. (i) A Riemannian space-time satisfying (5.8) is not Minkowskian in general.<sup>1)</sup>

(ii) If we use the identification (3.3) in place of (3.1), we have

$$F_{12}=F_{34}=0, \quad F_{14}=F_{31}=-\rho', \quad -F_{24}=F_{23}=\sigma' \quad (5.12)$$

in place of (5.9). (5.12) is of the same form as (5.9), for if we replace arbitrary functions  $\rho'$  and  $\sigma'$  by  $-\sigma$  and  $\rho$  respectively, (5.12) becomes (5.9).

## § 6. Comparison of both solutions

At first sight the exact solutions (GRS) and (NSS) explained in last section bear some resemblance to each other. If we compare them in detail, however, they differ remarkably in some points. We shall give an account of these circumstances in the following :

(i)  $F_{ij}$  and  $J^i$ .  $F_{ij}$ 's in both solutions are of the same form (5.1) and coincide with the corresponding one in the ordinary Maxwell theory. However, since both Riemannian space-times are not the same in general as will be seen in (ii) below,  $F_i^{\prime j}$ 's,  $F^{ij}$ 's and  $F_j^{\prime i}$ 's are not necessarily of the same forms.

Next we shall consider the charge-current vector  $J^i$ .  $J^i$ 's are zero in both solutions. But the circumstances concerning this vanishing of  $J^i$  are not the same. In solving (GRE) we have put  $J^i=0$  from the outset. This is seen in the second equation of (GRE). On the contrary, in solving (NSE) we obtained (NSS) without assuming  $J^i=0$ , and obtained it as a result. (See (5.11).)

Anyhow  $F_{ij}$ 's and  $J^i$ 's ( $=0$ ) are of the same forms in both solutions.

(ii) *Relation between  $F_{ij}$  and the Riemannian space-time.* First we shall consider (GRS). In this case  $g_{ij}$  and  $F_{ij}$  are connected by (5.2). When  $F_{ij}$  is given, (5.2) is a differential equation for  $g_{ij}$ , and conversely if  $g_{ij}$  is given, it becomes an algebraic equation concerning  $F_{ij}$ . Thus (5.2) indicates that the space-time and the electromagnetic field cannot be independent of each other. In other words, (5.2) shows that the electromagnetic field participates in the structure of the space-time through its energy

$E_{44} = \alpha(\rho, \sigma)/m$ . When and only when  $\rho = \sigma = 0$ , i. e. when the electromagnetic field is absent,  $g_{ij}$  becomes a solution of the purely gravitational equation for empty region.

If we consider the same problem in the case of (NSS) the situation is quite different from the above. In this case,  $h_{ij}$ , the quantity which determines the Riemannian space-time, is a solution of (5.8) which has no bearing on  $F_{ij}$ , and  $\rho$  and  $\sigma$  are quite arbitrary. There is no relation between  $h_{ij}$  and  $f_{ij}$ . This circumstance bears some resemblance to the case of the ordinary Maxwell theory which admits various plane waves in the same Minkowskian space-time. After all, gravitational waves and electromagnetic waves which are propagating in the same direction and are independent of each other can coexist in NS.

As was stated above,  $g_{ij}$  in (GRS) is a solution of (5.2) and  $h_{ij}$  of (NSS) is a solution of (5.8). Since the quadratic form  $\alpha(\rho, \sigma)$  is positive definite, both Riemannian space-time can never be the same so far as  $F_{ij} \neq 0$ . Moreover, both Riemannian space-times can never be the same no matter what  $\rho$ 's and  $\sigma$ 's we may take in both solutions.

(iii) *Superposition of the electromagnetic waves.* A remarkable feature of the ordinary Maxwell theory is that the field equations are linear and that it is possible to superpose any electromagnetic fields. On the contrary, both field equations (GRE) and (NSE) which we are dealing with are very complicated non-linear equations for  $F_{ij}$  and the fundamental tensor of the Riemannian space-times, and therefore we cannot expect any possibility of superposing electromagnetic fields in general.

In spite of these circumstances, however, we can show that the superposition of the electromagnetic fields is possible in some sense concerning the solution (NSS). To see this we take two solutions  $(h_{ij}; \rho_1, \sigma_1)$  and  $(h_{ij}; \rho_2, \sigma_2)$  belonging to (NSS) and having the same  $h_{ij}$  in common. If we put  $\rho_3 = c_1\rho_1 + c_2\rho_2$  and  $\sigma_3 = c_1\sigma_1 + c_2\sigma_2$ , where  $c_1$  and  $c_2$  are any constants, it seems most natural to interpret the words 'the superposition of the electromagnetic fields is possible' as that  $(h_{ij}; \rho_3, \sigma_3)$  satisfies (NSE) again. It is evident that this is the case with our (NSS). Accordingly, we can say in this sense that the superposition is possible.

In the case of (GRS), on the contrary, the situation is quite different. Let two solutions having the same  $g_{ij}$  in common be  $(g_{ij}; \rho_1, \sigma_1)$  and  $(g_{ij}; \rho_2, \sigma_2)$ . Then these solutions are connected by  $\alpha(\rho_1, \sigma_1) = \alpha(\rho_2, \sigma_2)$ . If  $\rho_3$  and  $\sigma_3$  are any linear combinations of  $\rho_1, \rho_2$  and  $\sigma_1, \sigma_2$  as given above, we have  $\alpha(\rho_3, \sigma_3) \neq \alpha(\rho_1, \sigma_1)$  in general. Hence  $(g_{ij}; \rho_3, \sigma_3)$  cannot be a solution of (GRE) and the superposition in the sense above stated is impossible. Moreover, it is difficult in GR to make the superposition possible no matter how we may modify its definition.

Summarizing the above, we can conclude that GR differs from NS considerably from the standpoint of the superposition of electromagnetic fields.

## § 7. Concluding remarks

We solved the field equations in GR and NS under the same conditions and by taking one of the simplest systems containing electromagnetic fields. Then we compared



the exact solutions thus obtained. Some different results as well as some common points were made clear.

The most remarkable and important result seems to be the one stated in (ii) of the last section. That is, the structure of the Riemannian space-time is closely connected with the electromagnetic field in GR, while in NS the structure is determined quite independently of the electromagnetic field. The former is quite natural. On the contrary, the latter seems to be somewhat strange. The curvature of the space-time is independent of the electromagnetic field. What has caused the curvedness of the space-time?

Our present research is only a simple consideration concerning a system of a very restricted type. Therefore it is difficult to come to some definite conclusion concerning the problem that which of the two theories is more appropriate to describe the system containing electromagnetic fields. The writer hopes, however, that the present research will turn out to be of some use in solving this problem.

The writer wishes to express his gratitude to Professor Y. Mimura and other members of the Institute for their helpful discussions.

## Appendix

It is not easy to solve (NSE) under only two assumptions 1 and 2. Therefore we put an additional assumption which we shall explain in the following.

It is known that  $F_{ij}$  defined by (3.1) satisfies as a consequence of (NSE),

$$F_{ij,k} + F_{jk,i} + F_{ki,j} = 0, \quad (\text{A} \cdot 1)$$

which is of the same form as the second half of the Maxwell equation given in (GRE). From Assumption 1, (A.1) and (4.4), we have

$$F_{12} = F_{34} = 0, \quad F_{14} = -F_{13} + a, \quad F_{24} = -F_{23} + b, \quad (\text{A} \cdot 2)$$

where  $a$  and  $b$  are arbitrary constants. If  $a$  or  $b$  is not zero, different from the case of the ordinary Maxwell equations or (GRE), the calculation to solve (NSE) becomes very complicated and it also becomes difficult to forecast the results. Because of this fact and the analogy with (GRS), we shall make the following assumption and shall deal with the simplest case:

Assumption 3:

$$a = b = 0. \quad (\text{A} \cdot 3)$$

As a consequence the electromagnetic field  $F_{ij}$  becomes of the same form as (5.1).

If we use this assumption we can prove (5.7), i. e.

$$f_{12} = f_{34} = f_{13} + f_{14} = f_{23} + f_{24} = 0. \quad (\text{A} \cdot 4)$$

*Proof.* It is known that the following relation holds for non-symmetric metric  $(g_{ij}^{(6),9})$

$$F_{ij} = \alpha f_{ij} + \beta \epsilon_{ijkl} f^{kl} / 2, \quad (\text{A} \cdot 5)$$

where  $f^{kl}$  is the tensor obtained by raising the indices of  $f_{ij}$  by means of  $h^{ij}$  which is defined by  $h_{ij} h^{jk} = \delta_i^k$ ,

$$\alpha = -p/\sqrt{-g}, \quad \beta = -h/\sqrt{-g}, \quad (\text{A} \cdot 6)$$

$$(g = \det. g_{ij}, \quad h = \det. h_{ij}; \quad p = \epsilon^{ijkl} f_{ij} f_{kl} / 8),$$

and  $p^2$  is equal to  $\det. f_{ij}$ .

We have from (A·2) and (A·3)

$$\begin{cases} \alpha f_{12} + \beta f_{34} = \alpha f_{34} + \beta f_{12} = 0, \\ \alpha (f_{13} + f_{14}) + \beta (f_{23} - f_{24}) = \alpha (f_{23} + f_{24}) - \beta (f_{13} - f_{14}) = 0. \end{cases} \quad (\text{A} \cdot 7)$$

Since the non-vanishing components of  $h^{ij} (= h^{ji})$  are

$$h^{11} = -B/m, \quad h^{12} = D/m, \quad h^{22} = -A/m, \quad -h^{33} = h^{44} = 1/C, \quad (\text{A} \cdot 8)$$

we have

$$\begin{cases} f^{12} = f_{12}/m, \quad f^{34} = -f_{34}/C^2, \\ f^{13} = (Bf_{13} - Df_{23})/mC, \quad f^{14} = -(Bf_{14} - Df_{24})/mC, \\ f^{23} = -(Df_{13} - Af_{23})/mC, \quad f^{24} = (Df_{14} - Af_{24})/mC. \end{cases} \quad (\text{A} \cdot 9)$$

Then (A·7) becomes

$$\begin{cases} \alpha f_{12} - \beta f_{34} C^2 = \beta f_{12} m - \alpha f_{34} = 0, \\ (\alpha - \beta D/mC) (f_{13} + f_{14}) + (\beta A/mC) (f_{23} + f_{24}) = 0, \\ (\beta B/mC) (f_{13} + f_{14}) - (\alpha + \beta D/mC) (f_{23} + f_{24}) = 0. \end{cases} \quad (\text{A} \cdot 10)$$

Since  $\beta$  cannot be zero,  $m \neq 0$ , and we are dealing with real quantities only, we can easily obtain (A·4) from these homogeneous equations for the components of  $f_{ij}$ .

Q. E. D.

From (A·4) we can deduce

$$p = 0, \text{ and accordingly } \det. f_{ij} = 0, \quad h = g, \quad (\text{A} \cdot 11)$$

which shows that our  $g_{ij}$  belongs to the third class in the sense of Hlavatý. One of the great obstacles which appear when we try to solve (NSE) is the fact that  $g$  and accordingly  $g^{ij}$  and  $h^{ij}$  become complicated. By the result above obtained, however, this obstacle is partly surmounted.

The remaining calculations to solve (NSE) is given in the reference.<sup>2)</sup>

Lastly we add a remark. If we start with the identification (3·3) in place of (3·1), the relation (A·1) does not necessarily hold. But if we assume (A·1), (A·4) is self-evident from Assumption 3.

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## The Magnetic Properties of the Praseodymium and the Neodymium Metals

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Along the same line on which we have previously discussed the cubic cerium metal, the magnetic properties of the succeeding rare earth metals, praseodymium and neodymium, are investigated. Because these metals have the structure of h. c. p., their crystalline field potentials cannot be uniquely determined so as to fit the experimental data. The analysis is, however, qualitatively satisfactory.

### § 1. Introduction

The magnetic properties of the rare earth metals have been discussed on the bases of Zener's mechanism for indirect exchange interaction<sup>1)</sup> and of the crystalline field potential<sup>2)</sup> for 4f electrons. The characteristic features of these metals are thought to be due to the simultaneous effects of them and have actually been understood to some extent from this point of view.

In the present paper we investigate the effect of the crystalline field on the magnetic properties of hexagonal rare earth metals. For this purpose we choose the praseodymium and the neodymium metals, because the effect of the crystalline field predominates the exchange field in these metals. The understanding of the crystalline field in these metals will be useful for further researches on complicated phenomena like ferromagnetic-antiferromagnetic transitions observed in other rare earth metals.

Pr, unlike Ce and Nd, has no antiferromagnetic phase.<sup>3),4)</sup> Pr shows a Shottky type anomaly in the atomic heat,<sup>3)</sup> whose maximum lies at a comparatively lower temperature than Ce and Nd. The magnetic susceptibility of Pr at low temperatures seems to tend to a finite value at 0°K.<sup>4)</sup> Nd has two anomalous peaks at 7.5°K and at 19°K<sup>3)</sup> besides the same kind of anomaly in specific heat as Pr. Both peaks may be regarded as the cooperative phase transitions and the former of them can be considered to correspond to the Néel point.<sup>5)</sup> The latter peak is suggested to be due to the interactions among the multipoles of the 4f electrons.<sup>6)</sup> The anisotropic magnetic susceptibility of the Nd single crystal has recently been reported.<sup>6)</sup> The electrical resistivity curves of Pr and Nd are concave downward over a wide temperature range and Nd shows another anomaly in the lower temperature range.<sup>7)</sup>

The phenomena mentioned above in the praseodymium and the neodymium metals can be explained semi-quantitatively by the view of the Stark splitting of the 4f electrons due to the crystalline field. We shall show in § 2 the calculation of the crystalline field potential, the results of which will be applied to Pr and Nd in § 3 and § 4, respectively.

In the present paper, we confine ourselves to analyzing the specific heat and the magnetic susceptibilities above the cooperative transition points. In § 5 discussion of our results will be made.

## § 2. Calculation of the crystalline field

While both metals, Pr and Nd, have the h. c. p. structure in the temperature range to which our interest will be confined, they have cubic closest packing structure at higher temperatures.<sup>5)</sup> Calculation of the crystalline field is performed on the model similar to that for the cerium metal which has already been reported by us<sup>1)</sup>: the valence electrons are treated as free electrons and smeared out uniformly over the whole of the metal, so that they have no contribution to the crystalline field discussed below. Therefore, the crystalline field can be obtained from the hexagonal closest packing of the ionized atoms.

The crystalline field potential for 4f electrons in the h. c. p. structure is expressed as

$$V = V_2^0 + V_4^0 + V_6^0 + V_6^6, \quad (1)$$

where

$$V_l^m = \sum_{\lambda} c_{lm} r_{\lambda}^l P_l^m(\cos \theta_{\lambda}) \cos m \varphi_{\lambda},$$

$$c_{l0} = \sum_n \frac{-ve^2}{R_n^{l+1}} P_l^0(\cos \theta_n), \quad (2)$$

$$c_{lm} = 2 \frac{(l-m)!}{(l+m)!} \sum_n \frac{-ve^2}{R_n^{l+1}} P_l^m(\cos \theta_n) \cos m \varphi_n.$$

In eq. (2),  $r_{\lambda}$ ,  $\theta_{\lambda}$  and  $\varphi_{\lambda}$  are the spherical coordinates of the  $\lambda$ -th electron in the atom whose nucleus is taken as the origin, and  $R_n$ ,  $\theta_n$  and  $\varphi_n$  the spherical coordinates of the nucleus of the  $n$ -th atom, and  $v$  the valency of the ionized atom. On account of the symmetry of the crystal structure the *sine* term does not appear in eq. (2).

In the lattice sum for  $l \geq 4$ , the direct sum method is applied over the range  $R_n \leq 4a$ , where  $a$  denotes the lattice constant along the  $a$ -axis. For  $l = 2$ , the convergency of the series of the direct sum is rather slow. We therefore adopted the Fourier method,<sup>6)</sup> whose convergency for  $l = 2$  is very rapid, and carried out for  $h_n \leq 4a$  in the reciprocal space. The calculation is performed for  $c/a = 1.579$  and  $1.613$  and the results are shown in Table 1 and Fig. 1. To verify we calculated  $c_{40}$  by the both methods for  $c/a = 1.613$ . In Table 1 cell,  $n$ , indicates the contribution from  $(n-1)a \leq R_n \leq na$  for direct summation and from  $na \leq h_n \leq (n+1)a$  for the Fourier method, except for  $n=1$  where  $h_n \leq 2/a$ . As is seen in Table 1, the larger  $l$  is, the more rapid the convergency is.

$c_{20}$  needs some comment. As can be seen in Fig. 1, the value of  $c_{20}$  is approximately zero when  $c/a = \sqrt{8/3} = 1.633$ . This is due to the symmetry of the lattice. When we carry out the lattice sum for  $c_{20}$ , the contributions cancel one another noticeably within the 1st cell and the total contribution from the 1st cell is reduced by a factor of  $10^2 \sim 10^3$ , which is just the order of magnitude to be cancelled by the contributions



Table 1. The calculated values of  $c_{lm}$ . ( $-ve^2/a^{l+1}$ ) should be multiplied to each value in the Table.  
 $c_{66} = c_{66}^* \times (2 \times 10395/12 !)$ .

$c/a$	Method	cell	$c_{20}$	$c_{40}$	$c_{60}$	$c_{66}^*$
1.579	Direct	1		+0.8355	-4.688	+5.704
		2		-0.2173	+0.196	-0.204
		3		-0.0142	+0.011	-0.025
		4		+0.0195	+0.003	-0.001
	sum	Total		+0.5845	-4.478	+5.474
	Fourier	1	+0.04799			
		2	+0.07960			
		3	+0.00155			
		Total	+0.12914			
1.613	Direct	1		+1.0508	-4.461	+5.753
		2		-0.2260	+0.174	-0.192
		3		-0.0159	+0.010	-0.009
		4		-0.0234	+0.003	-0.001
	sum	Total		+0.7855	-4.274	+5.551
	Fourier	1	-0.00598	+0.5689		
		2	+0.05754	+0.2159		
		3	+0.00152	-0.0062		
		Total	+0.05308	+0.7781		

from other cells. (cf. Table 1). The convergency of the lattice sum is nevertheless very rapid because the cancellations of the lattice sum in the 2nd and 3rd cells are less than those in the 1st cell and they show the characteristic convergency of the Fourier method. In the calculation of the energy levels of 4f-orbitals, the contribution from the term  $V_2^0$  is equally important as the other terms, and  $c_{20}$  is so small in magnitude and so sensitive to the value of  $c/a$  that it cannot have any rigorous meaning in view of our model used for the calculation of the crystalline field. In what follows, therefore, we modify  $D_{20}$  somewhat arbitrarily. (cf. eq. (3) and (4)).

Eq. (1) can be conveniently expressed in terms of equivalent operators as far as our interest is confined to the manifold of  $J^2 = \text{const.}$ ,<sup>10)</sup> i. e.,

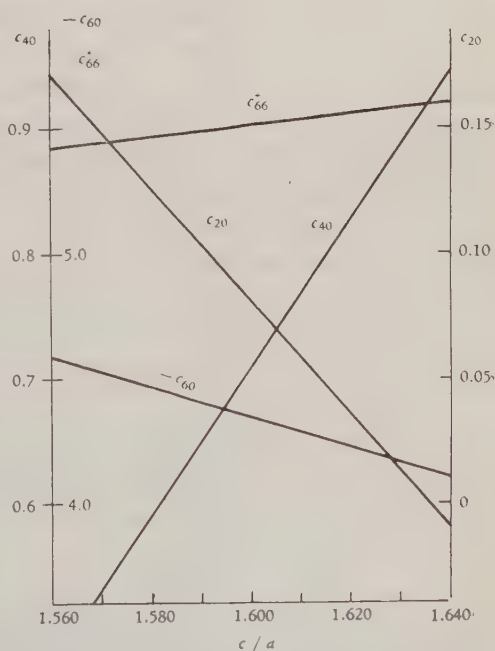


Fig. 1. The calculated values of  $c_{lm}$ . ( $-ve^2/a^{l+1}$ ) should be multiplied to each value of the ordinate.

$$V = D_{20}J_{20} + D_{40}J_{40} + D_{60}J_{60} + D_{66}J_{66}, \quad (3)$$

where

$$\begin{aligned} J_{20} &= 3J_z^2 - J(J+1), \\ J_{40} &= 35J_z^4 - 30J(J+1)J_z^2 + 25J_z^2 - 6J(J-1) - 3J^2(J-1)^2, \\ J_{60} &= 231J_z^6 - 315J(J+1)J_z^4 + 735J_z^4 + 105J^2(J+1)^2J_z^2 \\ &\quad - 525J(J+1)J_z^2 + 294J_z^2 - 5J^3(J+1)^3 + 40J^2(J+1)^2 \\ &\quad - 60J(J+1), \\ J_{66} &= \frac{1}{2}[(J_x + iJ_y)^6 + (J_x - iJ_y)^6], \\ D_{lm} &= r^l \alpha_{lm} c_{lm} \times (\text{coefficient of } P_l^m) \end{aligned} \quad (4)$$

In eq. (4),  $\alpha_{20}$ , for example, corresponds to the  $\alpha$  in Stevens' notations,<sup>10</sup> and  $r$  is also due to his notation.

### § 3. Praseodymium

The most stable ionic state of Pr is  $\text{Pr}^{3+}$  whose ground state is  ${}^3\text{H}_4$ . The next excited state is  ${}^3\text{H}_5$  which is estimated theoretically to be higher by  $2000 \text{ cm}^{-1}$  than the ground state.<sup>11)</sup> Therefore, we discard the possibility of  $\text{Pr}^{2+}$  being in the  $\text{H}$  state. The splitting of the energy level,  ${}^3\text{H}_4$ , by the crystalline field (3) can be obtained in the same way as in the case of  $\text{Ce}^{3+}$ : In the present case, however, the difficulty arises from the fact that the relative magnitude of  $r^l$ , where  $l$  takes 2, 4 and 6, cannot be determined uniquely. There is no available calculation for the  $4f$  wave functions in the rare earth atoms. We therefore employ for the radial part of the  $4f$  wave function the Slater function containing the effective nuclear charge  $Z^*$  as a parameter which will be determined below. In Fig. 2, the energy levels are shown as a function of  $Z^*$  for the observed values of  $a$  ( $= 3.662 \text{ \AA}$ ) and  $c$  ( $= 1.613$ ).<sup>12)</sup> The  $\uparrow$ 's in the figure indicate the following eigenfunctions.

$$\begin{aligned} |A1\rangle &= N_{A1}(|\pm 4\rangle + C_{A1}|\mp 2\rangle) && (\text{doublet}), \\ |A2\rangle &= N_{A2}(|\pm 4\rangle + C_{A2}|\mp 2\rangle) && (\text{doublet}), \\ |B1\rangle &= B_{B1}(|+3\rangle + C_{B1}|-3\rangle) && (\text{singlet}), \\ |B2\rangle &= N_{B2}(|+3\rangle + C_{B2}|-3\rangle) && (\text{singlet}), \\ |C\rangle &= |\pm 1\rangle && (\text{doublet}), \\ |D\rangle &= |0\rangle && (\text{singlet}). \end{aligned} \quad (5)$$

In eq. (5), the state  $|+4\rangle$ , for example, represents the normalized wave function of  $J_z = +4$ .  $N_i$  is the normalization constant and is equal to  $(1 + C_i^2)^{-1/2}$ , where  $i$  stands for  $A$  or  $B$  and  $i$  for 1 or 2. As can be seen from eq. (4), only the states

that are different from each other by 6 in the value of  $J_z$  can combine together, and the upper (or lower) signs of the former double signs in the right-hand sides of eq. (5) correspond to the upper (or lower) signs of the latter ones. The possible ground state is either  $|D\rangle$  or  $|B1\rangle$  depending on  $Z^*$ . In either case, that the ground state of  $\text{Pr}^{3+}$  is a singlet explains the fact that the praseodymium metal has no antiferromagnetic phase. Furthermore, the levels crowd relatively near the ground state and this fact is also consistent with the fact that the Shottky type anomaly of the atomic heat appears in a comparatively lower temperature range than in the cases of the cerium and the neodymium metals.

With these theoretical backgrounds, our calculations are carried out for the atomic heat and the magnetic susceptibility. The fair agreements with the observations are obtained for  $Z^*=10.5$  with  $D_{20}$  multiplied by 2. The multiplication factor is necessary, for without such a modification for  $D_{20}$ , the results are not good regardless of the choice of  $Z^*$ . In Table 2, we show the energy eigenvalues corresponding to the eigenfunctions in eq. (5) together with coefficients  $C_{\delta i}$  in the same equation. The eigenvalue of the ground state is taken as zero.

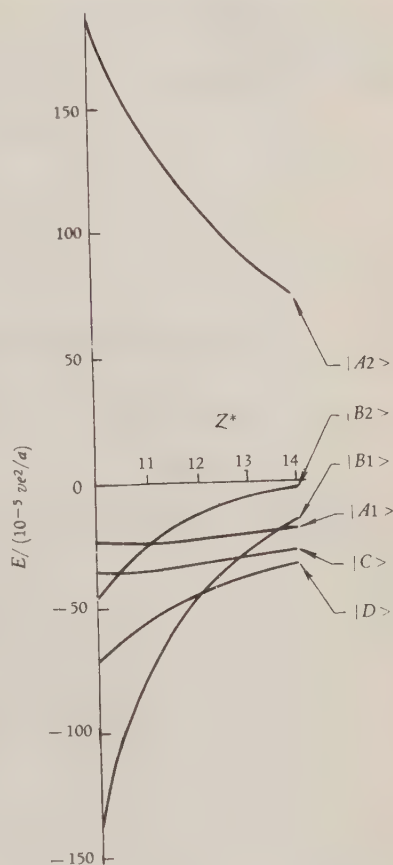


Fig. 2. The energy levels of the 4f electrons of  $\text{Pr}^{3+}$  ( $^3\text{H}_4$ ) in the crystal-line field as a function of  $Z^*$ .

Table 2. The energy eigenvalues and the coefficients of the eigenfunctions of the 4f electrons in the Pr metal.

	$ A1\rangle$	$ A2\rangle$	$ B1\rangle$	$ B2\rangle$	$ C\rangle$	$ D\rangle$
(Energy eigenvalue)/( $10^{-5} ve^2/a$ )	84.3	400	60.4	132	36.4	0
$C_{\delta i}$	11.6	-0.0865	1	-1		

By using the energy eigenvalues,  $E_i$ , given in Table 2 and the multiplicity,  $n_i$ , shown in eq. (5), we can calculate the atomic heat due to the localized 4f-electrons from the following equation :

$$\Delta C = Nkx^2 \left\{ (1/Z_0) (\sum_i n_i E_i^2 e^{-E_i x} - (1/Z_0)^2 (\sum_i n_i E_i e^{-E_i x})^2 \right\}, \quad (6)$$

where  $Z_0 = \sum_i n_i e^{-x E_i}$ .



In above equations  $E_i$ 's express the numerical values given in Table 2 (i. e. the value measured in the unit of  $(10^{-5} \text{ } \nu e^2/a)$ ) and  $x=10^{-5} (\text{ } \nu e^2/a) kT$ . The magnetic susceptibilities are calculated in the same way as in the case of  $\text{Ce}^{2+}$  and expressed as

$$\begin{aligned}\chi_{\parallel} &= \frac{N(g\mu_B)^2}{kT} \frac{1}{Z_0} \left( F_{\parallel} + \frac{1}{x} G_{\parallel} \right), \\ \chi_{\perp} &= \frac{N(g\mu_B)^2}{kT} \frac{1}{Z_0} \left( F_{\perp} + \frac{1}{x} G_{\perp} \right),\end{aligned}\quad (7)$$

where

$$\begin{aligned}F_{\parallel} &= 0.000444 (17240 e^{-84.3x} + 70540 e^{-400x}) + 2e^{-36.4x}, \\ G_{\parallel} &= 1.43 \frac{e^{-84.3x} - e^{-400x}}{316} + 18 \frac{e^{-60.4x} - e^{-36.4x}}{71.8}, \\ F_{\perp} &= 0, \\ G_{\perp} &= 0.0149 \left\{ 331 \frac{e^{-102x} - e^{-400x}}{268} + 408 \frac{e^{-84.3x} - e^{-102x}}{47.9} \right. \\ &\quad + 209 \frac{e^{-60.4x} - e^{-400x}}{340} + 530 \frac{e^{-60.4x} - e^{-84.3x}}{23.9} + 9 \frac{e^{-36.4x} - e^{-400x}}{364} \\ &\quad \left. + 1200 \frac{e^{-36.4x} - e^{-84.3x}}{47.9} - 20 \frac{1 - e^{-36.4x}}{36.4} \right\},\end{aligned}\quad (8)$$

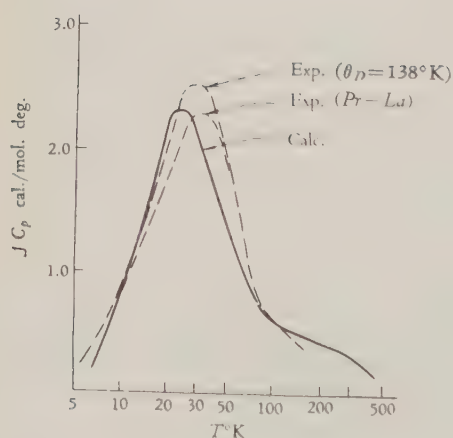


Fig. 3. The part of the atomic heat due to the 4f electrons of Pr metal. The calculated curve is shown together with the experimental ones.

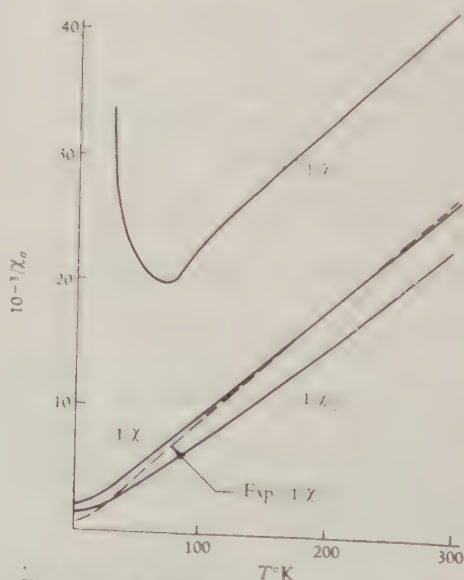


Fig. 4. The magnetic susceptibilities calculated for the Pr metal are shown together with the experimental curve.

In these equations,  $N$  is the number of atoms,  $g$  the Lande  $g$ -factor equal to  $4/5$ ,  $\mu_B$  the Bohr magneton, and  $Z_0$  the partition function.  $\chi_{\parallel}$  means the susceptibility when the external magnetic field is parallel to the  $c$ -axis and  $\chi_{\perp}$  the one when the magnetic field is perpendicular to the same axis.  $F_{\parallel}$  and  $F_{\perp}$  are the contributions from the matrix elements between degenerate states, and  $G_{\parallel}$  and  $G_{\perp}$  are those from the matrix elements between non-degenerate states. The exchange effect is omitted here. The results are shown in Figs. 3 and 4 together with the experimental data. In the actual calculation  $v$  is taken as 3. In Fig. 3, the experimental curve indicated by (Pr-La) means the excess part of Pr atomic heat over La's.<sup>3)</sup> The other experimental curve is the one obtained with the Debye temperature  $\theta_D = 138^\circ\text{K}$ .<sup>13)</sup> In Fig. 4  $\chi = \chi_{\parallel}/3 + 2\chi_{\perp}/3$ , which should be compared with the experimental data.  $\chi_{\parallel}$  and  $\chi_{\perp}$  are very sensitive to the character of the ground state but there is no observation to be referred to.

#### § 4. Neodymium

We treat the neodymium metal in the similar way as for the praseodymium metal. The ground state is  $^4I_{9/2}$  and the next excited state,  $^4I_{11/2}$ , lies higher by  $1800\text{ cm}^{-1}$ .<sup>11)</sup> Therefore, only  $J=9/2$  manifold is considered. The observed values of  $c/a$  are  $1.613$ <sup>12)</sup> and  $1.609$ ,<sup>14)</sup>

but the former is taken here with the value of  $a = 3.650\text{ \AA}$ . For Nd, there remains Kramers' doublet on account of the odd number of the  $4f$  electrons, and all levels are doublets as is shown in Fig. 5. The notations in the figure are similar to the case of Pr. Those eigenstates are represented by the following expressions:

$$\begin{aligned}
 |A1\rangle &= N_{A1}(|\pm 9/2\rangle + C_{A1}|\pm 3/2\rangle) & (\text{doublet}), \\
 |A2\rangle &= N_{A2}(|\pm 9/2\rangle + C_{A2}|\mp 3/2\rangle) & (\text{doublet}), \\
 |B1\rangle &= N_{B1}(|\pm 7/2\rangle + C_{B1}|\mp 5/2\rangle) & (\text{doublet}), \\
 |B2\rangle &= N_{B2}(|\pm 7/2\rangle + C_{B2}|\mp 5/2\rangle) & (\text{doublet}), \\
 |C\rangle &= |\pm 1/2\rangle & (\text{doublet}),
 \end{aligned} \tag{9}$$

where similar notations have been used as in eq. (5).

The calculations have been performed for  $Z^* = 11.0$  with  $D_{20}$  multiplied by a factor

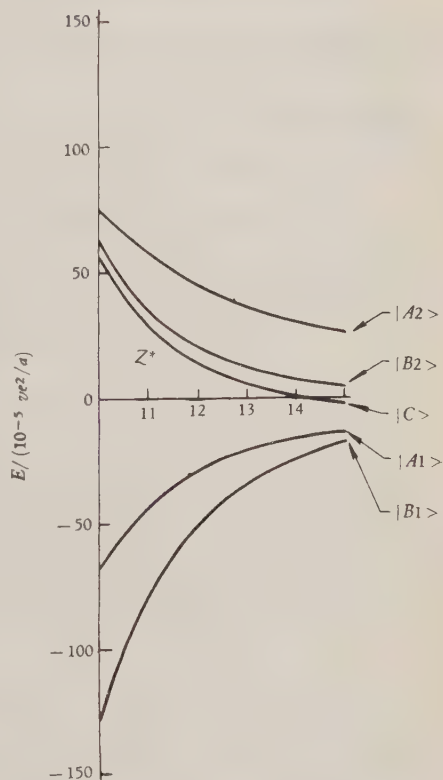


Fig. 5. The energy levels of the  $4f$  electrons of  $\text{Nd}^{3+}$  ( $^4I_{9/2}$ ) in the crystalline field.

2.5. The coefficients in eq. (9) and the energy eigenvalues are listed in Table 3. The zero of energy is set on the ground state.

Table 3. The energy eigenvalues and the coefficients of the eigenfunctions of the 4*f* electrons in the Nd metal.

	$ A1\rangle$	$ A2\rangle$	$ B1\rangle$	$ B2\rangle$	$ C\rangle$
(Energy eigenvalue)/(10 <sup>-5</sup> <i>ve</i> <sup>2</sup> / <i>a</i> )	16.7	197.9	0	137.8	76.7
<i>C</i> <sub>8<i>l</i></sub>	-7.85	0.127	-3.70	0.270	

In this case, the specific heat of the Shottky type can also be calculated by eq. (6) with *E<sub>i</sub>*'s in Table 3 and *n<sub>i</sub>*'s in eq. (9). The magnetic susceptibilities are expressed by eq. (7) with *g* equal to 8/11 and with *F*'s and *G*'s given by the following :

$$F_{\parallel}=8.77+19.1e^{-137.8x}+3.94e^{-16.7x}+38.7e^{-197.9x}+0.5e^{-76.7x},$$
$$G_{\parallel}=9.14\frac{1-e^{-137.8x}}{137.8}+2.26\frac{e^{-16.7x}-e^{-197.9x}}{181.2},$$
$$F_{\perp}=2.03(1+e^{-137.8x})+12.5e^{-76.7x},$$
$$G_{\perp}=20.1\frac{1-e^{-16.7x}}{16.7}+12.0\frac{1-e^{-137.8x}}{137.8}-0.04\frac{1-e^{-197.9x}}{197.9}$$
$$+0.675\frac{e^{-16.7x}-e^{-137.8x}}{121.1}+23.6\frac{e^{-16.7x}-e^{-76.7x}}{60.6}$$
$$+0.383\frac{e^{-76.7x}-e^{-197.9x}}{121.2}+9.15\frac{e^{-16.7x}-e^{-197.9x}}{60.1}.$$

(10)

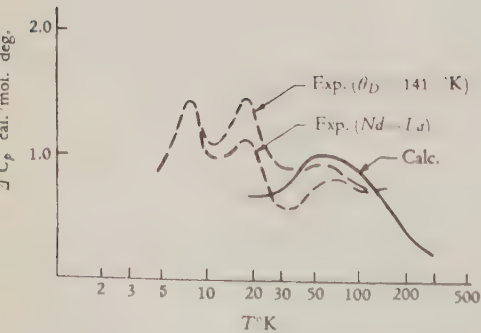


Fig. 6. The part of the atomic heat due to the 4*f* electrons of Nd metal. The calculated curve is shown together with the experimental ones.

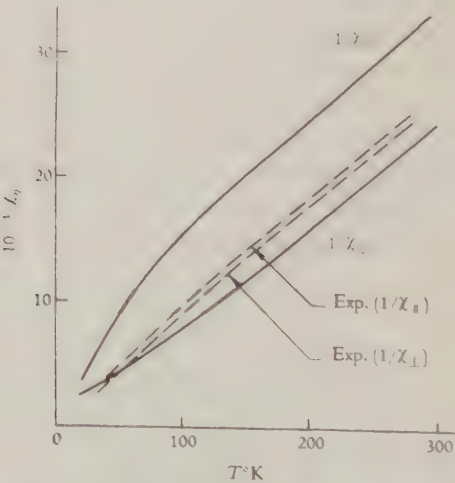


Fig. 7. The magnetic susceptibilities calculated for the Nd metal are shown together with the experimental curves.



The results are shown in Fig. 6 and 7, where  $v=3$  is also taken and the notations are similar to those in the case of Pr. Again the exchange interaction is not taken into account.

As is seen from Fig. 5, there is one doublet near the ground state and this situation does not change even if some modification of  $D_{20}$  is made. This explains the experimental fact that the Nd metal shows  $\Delta S = R \ln 4$  below the second cooperative transition point which lies at a comparatively low temperature.<sup>3)</sup> In Table 2, we show the excess entropy corresponding to the anomalous atomic heat,  $\Delta C_p$ . In the last column there are also shown the theoretical values to be compared with. As has been discussed in § 1, the cooperative transition points are at 7.5°K and 19°K.

Table 4. The entropies related to the excess atomic heats of the Nd metal.

$T^\circ\text{K}$	$\Delta S(\text{Nd}-\text{La})$	$\Delta S(\theta_D=141^\circ\text{K})$	
10	1.18	1.2	$R \ln 2 = 1.38$
25	2.02	2.2	$R \ln 4 = 2.75$

The magnetic susceptibility has been measured for both single<sup>6)</sup> and poly-crystals.<sup>4),15)</sup> The observed anisotropy of  $\chi$  is smaller than the result obtained here, but it is qualitatively understood that  $\chi_\perp$  is larger than  $\chi_\parallel$  and that the sudden change of the slope of the experimental  $1/\chi - T$  curve is probably related to the effect of the crystalline field.

## § 5. Discussion

So far we have used the three parameters,  $v$ ,  $Z^*$  and  $D_{20}$ .  $v$  determines only the absolute scale of energy as can be seen from Figs. 2 and 5.  $v=3$  seems to be somewhat large compared with the case of the cerium metal where  $v=2.5$  was consistent.<sup>2)</sup> But it would not be unreasonable. The values of  $Z^*$ , 10.5 and 11.0, should be compared with 12.65 and 13.3 obtained from Slater's rule for the Pr and the Nd atoms, respectively. Some discrepancy between the Slater function and the wave function calculated numerically by the Hartree method can also be found for the  $4f$  wave functions of Au and Tl atom.<sup>16)</sup> It is difficult to make clear the meaning of the modification for the  $D_{20}$  on the basis of the present model of the crystalline field. We can probably say that a small departure from our model for crystalline field may be responsible for this. The results obtained in this paper should be understood to be rather qualitative or semi-quantitative. It cannot be uniquely determined whether the ground state of Pr is  $|D\rangle$  or  $|B1\rangle$ . A better agreement with the observations can be obtained if we choose  $|D\rangle$  as the ground state and the realization with proper  $z^*$  and  $v$  relates to the modification as  $D_{20}$ . To explain the susceptibility and the specific heat by taking  $|B1\rangle$  as the ground state, a fairly large exchange interaction is necessary. A large exchange effect is not plausible, so that this situation is abandoned in this paper.

The discrepancies between the present analysis and the experimental data can be related to the following facts:

- (1) The modified hexagonal structure has been reported.<sup>12),17)</sup>
- (2) The temperature dependency of  $c/a$  has not yet been observed, which sensitively affects  $D_{20}$ .
- (3) The conduction electrons may deviate from the free electrons and the polarization of the core electrons, especially those in  $5s$  and  $5p$  orbitals, may arise due to the  $4f$  electrons.
- (4) There is a lack of our knowledge about the  $4f$  wave function in the metal. It is possible that there is a marked deviation from the atomic or Slater's function.

For example, the larger anisotropy of the susceptibility for the Nd metal which is obtained in the present calculation would be intuitively understood in view of (1) and (3) mentioned above, because these factors have a tendency to mix the levels further than those indicated in Figs. 2 and 5 and diminish the anisotropy. Detailed considerations on these points will be made in the future.

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## Parity Nonconservation and the Beta-Spectrum of RaE\*

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A method suitable for analysing the anomalous  $\beta$ -spectrum of RaE is developed. It is shown that, if the Gamow-Teller component of the  $\beta$ -decay interaction is of the tensor type, it is very difficult to explain the shape of the  $\beta$ -spectrum of RaE unless all of the following conditions are satisfied simultaneously: (a) the Fermi component is of the scalar type, (b)  $C_S/C_S' = C_T/C_T'$ , and (c)  $C_S/C_T = \text{real}$ . Alternatively, if the Gamow-Teller component is of the axial vector type, the conditions are: (a) the Fermi component is of the vector type, (b)  $C_V/C_V' = C_A/C_A'$ , and (c)  $C_V/C_A = \text{real}$ .

### § 1. Introduction and summary

Since the experiments by Wu et al<sup>1)</sup> showed the violence of the space reversal invariance all our previous knowledge about the type of the  $\beta$ -decay interaction has been undergoing a thorough re-investigation. Although many recent data on angular distribution of  $\beta$ -rays from polarized nuclei, longitudinal polarization of  $\beta$ -rays, and angular correlations between  $\beta$ -rays and circularly polarized  $\gamma$ -rays, etc. (except for the data on electron-neutrino angular correlations), gave us a fairly consistent picture of the interaction type, they are still not sufficient to determine it definitively.

Though the peculiar  $\beta$ -spectrum of RaE is seemingly difficult to explain, it is now well-known that it can be reasonably explained as an accidental case in which considerable cancellations occur among several partial matrix elements<sup>2)</sup>. The aim of this paper is to re-investigate the cancellation spectrum by taking into account the fact of parity nonconservation in  $\beta$ -decay and to obtain additional information about the type of the  $\beta$ -decay interaction. A similar analysis was made by Lewis<sup>3)</sup>.

Through out this paper the following assumptions are made: 1) The Fermi theory<sup>4)</sup> in a broad sense is valid, in which the four spinor fields interact at a point without derivative coupling<sup>5)</sup>. Parity nonconservation is taken into account.<sup>6)</sup>

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2) The time reversal invariance of the Wigner type<sup>7)</sup> is valid for strong interactions, so that the relative phases of nuclear matrix elements can be fixed<sup>7)</sup>, while no such assumption is made for the weak interactions<sup>9)</sup>.

If the parity nonconservation is taken into account, the interaction Hamiltonian for  $\beta$ -decay can be written as a sum of two parts, one with coupling constants  $C_i$ 's and the other with  $C_j$ 's in the notation of Lee and Yang<sup>6)</sup>, where  $i, j=S, V, T, A$  and  $P$ . For  $\beta$ -spectra there exists no interference term between these two parts. Therefore, the only quantities which can give us important knowledge about the validity of the two-component neutrino theory<sup>10)</sup> and the time reversal invariance from study of  $\beta$ -spectra are

$$\alpha_{ij} = \Re(C_i^* C_j + C_i'^* C_j') - (C_i^2 + C_i'^2)(C_j^2 + C_j'^2)^{1/2}. \quad (1)$$

Here  $\Re$  means the real part.  $\alpha_{ij}$  is always equal to or smaller than unity. If we put  $i=S(T)$  and  $j=V(A)$  in (1), the  $\alpha_{ij}$  appears as the coefficient of the Fierz term<sup>11)</sup>. The empirical fact that the Kurie plots of the allowed spectra are nearly straight can be explained by the Fierz conditions,

$$\alpha_{SV}=0, \quad \alpha_{TA}=0. \quad (2)$$

On the other hand we can obtain other conditions from cancellation spectra of forbidden transitions such as the decays of RaE and  $C_{40}^{187}$ , because in these cases the interference terms involving  $\alpha_{SV}$  and  $\alpha_{TA}$  play the most important parts. By the analysis of RaE spectrum we are led to the conclusion that the peculiar shape can be explained only by the optimum effect of the interference terms and even a small deviation from the optimum situation makes the explanation remarkably difficult. The optimum situation is realized by the following conditions:

$$\begin{aligned} C_S/C_S' &= C_T/C_T', \quad C_V/C_V' = C_A/C_A', \\ C_S/C_T &= \text{real}, \quad C_V/C_A = \text{real}. \end{aligned} \quad (3)$$

## § 2. Theoretical expression for the correction factor

Deviations of  $\beta$ -spectra from the allowed shape are expressed by correction factors introduced by Konopinski and Uhlenbeck<sup>12)</sup>. Though their formulation is useful, several corrections must be taken into account in order to make it more accurate. Among them the effects due to the finiteness of nuclear size are considered to be the most important for RaE decay. They are composed of the finite nuclear size effect in the narrow sense<sup>13)</sup>, the finite de Broglie wave length effect<sup>14)</sup>, and the effect of the  $\beta$ -decay from inside of the nucleus<sup>15)</sup>. These effects were originally proposed independently of each other. Takebe<sup>16)</sup> proposed a general formulation including the above corrections synthetically, but the number of unknown parameters seems too large for practical purposes.

Recently two of the authors<sup>17)</sup> proposed a formulation convenient for practical uses in analysing  $\beta$ -spectra taking into account the above mentioned effects and the screening effect<sup>18)</sup> synthetically. It was shown that the effects due to finite nuclear size can be treated without detailed knowledge about the nuclear charge distribution if it is spherically sym-

metric, so far as we regard the nuclear matrix elements as unknown parameters. Namely the main part of this correction can be included legitimately by only replacing the ordinary nuclear radius in the Konopinski-Uhlenbeck<sup>(12)</sup> formulas for  $\alpha Z \ll 1$  ( $\alpha$ : fine structure constant,  $Z$ : atomic number) by the "effective nuclear radii", which are expected to be about the same as or somewhat larger than the ordinary one. Moreover, the unimportance of the screening effect on the RaE spectrum (except for the very low energy region of  $W \lesssim 1.1$ ) is suggested even if the cancellation is remarkable.

According to this theory the general correction factor for the RaE spectrum (spin change  $1^- \rightarrow 0^+$ ),  $C_{1SVTA}(W)$  is given as follows:

$$C_{1SVTA}(W) = C_{1SVTA}^{(a)}(W) + C_{1SVTA}^{(b)}(W), \quad (4)$$

with

$$\begin{aligned} C_{1SVTA}^{(a)}(W) = & |C_T| \left[ \left( \int \beta \mathbf{a} \cdot \mathbf{I}_1(r) \right) - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right] \\ & - i \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} C_S \left( \int \beta \mathbf{r} \cdot \mathbf{I}_1(r) \right) \Big|^2 \\ & + \frac{1}{3} \left( q - \frac{p^2}{W} \right) \left[ \left( i C_S \int \beta \mathbf{r} \cdot \mathbf{I}_1(r) \right)^* \left\{ C_T \left[ \left( \int \beta \mathbf{a} \cdot \mathbf{I}_1(r) \right) \right. \right. \right. \\ & \left. \left. - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right] - i \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} C_S \left( \int \beta \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right\} + c.c. \right] \\ & - \frac{1}{3} \left( q + \frac{p^2}{W} \right) \left[ \left( C_T \int \beta \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right)^* \left\{ C_T \left[ \left( \int \beta \mathbf{a} \cdot \mathbf{I}_1(r) \right) \right. \right. \right. \\ & \left. \left. - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right] - i \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} C_S \left( \int \beta \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right\} + c.c. \right] \\ & + \left( \frac{q^2 + p^2}{3} - \frac{2qp^2}{9W} \right) |C_S \int \beta \mathbf{r} \cdot \mathbf{I}_1(r)|^2 + \left( \frac{q^2 + p^2}{6} + \frac{2qp^2}{9W} \right) |C_T \int \beta \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r)|^2 \\ & + |C_V \left( \int \mathbf{a} \cdot \mathbf{I}_1(r) \right) - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) \\ & \quad - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} C_V \left( \int \mathbf{r} \cdot \mathbf{I}_1(r) \right)|^2 \\ & - \frac{1}{3} \left( q + \frac{p^2}{W} \right) \left[ \left( i C_V \int \mathbf{r} \cdot \mathbf{I}_1(r) \right)^* \left\{ C_V \left( \int \mathbf{a} \cdot \mathbf{I}_1(r) \right) \right. \right. \right. \\ & \left. \left. - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} C_V \left( \int \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right\} + c.c. \right] \\ & + \frac{1}{3} \left( q - \frac{p^2}{W} \right) \left[ \left( C_A \int \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right)^* \left\{ C_V \left( \int \mathbf{a} \cdot \mathbf{I}_1(r) \right) \right. \right. \right. \\ & \left. \left. - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} \cdot \mathbf{I}_1(r) \right) - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} C_V \left( \int \mathbf{r} \cdot \mathbf{I}_1(r) \right) \right\} + c.c. \right] \end{aligned}$$

$$\begin{aligned}
& + \left( \frac{q^2 + p^2}{3} + \frac{2qp^2}{9W} \right) C_V \int \mathbf{r} I_1(r) - \left( \frac{q^2 - p^2}{6} - \frac{2qp^2}{9W} \right) C_A \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \\
& + \frac{1}{W} \left\{ \left[ C_V \left( \int \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right. \right. \\
& \quad \left. \left. - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} C_V \left( \int \mathbf{r} I_1(r) \right) \right] * \right. \\
& \quad \times \left\{ C_T \left[ \left( \int \beta \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right] \right. \\
& \quad \left. \left. - i \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} C_S \left( \int \beta \mathbf{r} I_1(r) \right) \right\} \right. \\
& + \frac{q}{3} \left\{ i C_S \left( \int \beta \mathbf{r} I_1(r) \right) - C_T \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right\} * \\
& \quad \times \left\{ C_V \left( \int \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right. \\
& \quad \left. \left. - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} C_V \left( \int \mathbf{r} I_1(r) \right) \right\} \right. \\
& - \frac{q}{3} \left\{ i C_V \left( \int \mathbf{r} I_1(r) \right) - C_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right\} * \\
& \quad \times \left\{ C_T \left[ \left( \int \beta \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right] \right. \\
& \quad \left. \left. - i \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} C_S \left( \int \beta \mathbf{r} I_1(r) \right) \right\} \right. \\
& - \frac{q^2 + p^2}{3} (i C_V \int \mathbf{r} I_1(r)) * (i C_S \int \beta \mathbf{r} I_1(r)) \\
& - \frac{q^2 + p^2}{6} (C_A \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r)) * (C_T \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r)) + c.c. \Big\}, \quad (5)
\end{aligned}$$

and  $C_{NSR}^{(b)}(W)$  the expression (5) with all the coupling constants replaced by primed ones,  $C'_i$ . In (5), c.c. means complex conjugate,  $p$  and  $q$  represent the electron and neutrino momenta respectively, and  $W$  the electron energy including its rest mass. We

\* These approximations correspond to the omission of  $\alpha Z W \rho$  in comparison with unity.  $F_1(\rho)$  and  $I_2(r)$  are included only in the  $L_1$  term<sup>12,17)</sup> which has no importance in RaE decay. Although the approximation  $I_2(r) = I_1(r)$  reduces the number of nuclear matrix elements by one, it is reasonably permitted because of the absence of interference between the  $L_1$  term and the main term. The approximation of,  $\lambda^+ = \mu^+ = \nu^+ = 1$  and  $\nu^- = 0$  concerns with the main term, but does not reduce the number of nuclear matrix elements. Thus the effect of this approximation is considerably smaller than the effect of the new nuclear matrix elements which will be discussed in § 4 as neglected terms in the effective radii theory.



use the natural units ( $\hbar=c=1$ ) and also take the electron mass as unity ( $m=1$ ). The first forbidden transition including only scalar, tensor and pseudoscalar types is explicitly treated in ref. 17. Expression (5) is obtained by the general prescription in ref. 17 with additional approximations of  $F_1(\rho)/F_0(\rho)=1$ ,  $I_2(r)=I_1(r)$ ,  $\lambda^\pm=\mu^\pm=\nu^\pm=1$  and  $\nu^-=0$  in the notation of ref. 17.\* We can improve (4) by removing these additional approximations, but the degree of improvement is so small and insignificant for the following argument that we use (5) in this paper.

Each term of (5) clearly corresponds to that of the familiar Konopinski-Uhlenbeck formula for low  $Z$ . The energy independent function  $I_1(r)$  is defined by eq. (13) of ref. 17, and its behaviors in nuclei are shown in Figs. 3 to 6 of ref. 17. The effective nuclear radii,  $\rho_1(X)$ 's are defined by eq. (21) of ref. 17 as the ratios of nuclear matrix elements. Different  $\rho_1$ 's have the same order of magnitude but may take different values. Their magnitudes are comparable with that of the ordinary nuclear radius and are likely to be somewhat larger. Since the exact values of the nuclear matrix elements are not known, we need to treat the effective nuclear radii as flexible quantities to some extent.

It is convenient in our analysis to expand eq. (4) into a finite power series in the electron energy  $W$  by using the relations,  $q=W_0-W$  ( $W_0$ : the maximum electron energy and  $p^2=W^2-1$ ).

$$C_{ISVTA}(W) = a_{-1}/W + a_0 + a_1W + a_2W^2, \quad (6)$$

where  $a_i$ 's are energy independent coefficients.

The explicit forms of  $a_1$  and  $a_2$  are

$$\begin{aligned} a_1 = & \frac{4}{3} \left\{ \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} - \frac{2}{3} W_0 \right\} \varepsilon_s^2 \left| \int \beta \mathbf{r} I_1(r) \right|^2 \\ & + \left\{ \frac{4}{3} \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} - \frac{5}{9} W_0 \right\} \varepsilon_A^2 \left| \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2 \\ & - \frac{4}{3} \alpha_{ST} \varepsilon_S \varepsilon_T (i \int \beta \mathbf{r} I_1(r)) * \left\{ \left( \int \beta \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right\} \\ & - \frac{4}{3} \alpha_{VA} \varepsilon_V \varepsilon_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) * \left\{ \left( \int a I_1(r) \right) - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} \left( \int \mathbf{r} I_1(r) \right) \right\} \\ & - \frac{4}{3} \alpha_{SV} \varepsilon_S \varepsilon_V (i \int \mathbf{r} I_1(r)) * (i \int \beta \mathbf{r} I_1(r)) \\ & - \frac{2}{3} \alpha_{TA} \varepsilon_T \varepsilon_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) * \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \\ & - \frac{1}{9} W_0 \varepsilon_T^2 \left| \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2 - \frac{4}{9} W_0 \varepsilon_V^2 \left| \int \mathbf{r} I_1(r) \right|^2, \end{aligned} \quad (7)$$

and

$$a_2 = \frac{8}{9} \varepsilon_s^2 \left| \int \beta \mathbf{r} I_1(r) \right|^2 + \frac{1}{9} \varepsilon_T^2 \left| \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2$$

$$+\frac{4}{9}\varepsilon^2\nu|\int\mathbf{r}I_1(r)|^2+\frac{5}{9}\varepsilon_A^2|\int\boldsymbol{\sigma}\times\mathbf{r}I_1(r)|^2, \quad (8)$$

where we defined  $\varepsilon_i$  as  $\varepsilon_i=(|C_i|^2+|C'_i|^2)^{1/2}$ .

It is interesting that  $a_2$  is always positive,  $a_2>0$ .

If we assume the Fierz condition (2), (6) reduces to a simpler form.

### § 3. Analyses of the experimental data

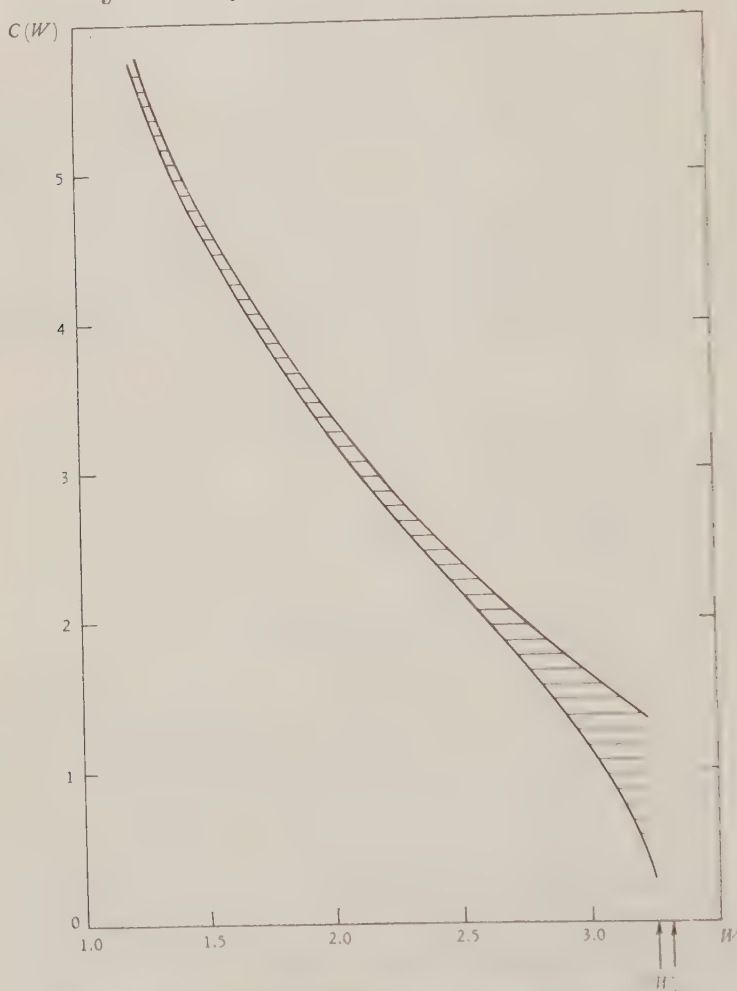


Fig. 1. Experimental correction factor for the RaE  $\beta$ -spectrum obtained by Plassman and Langer<sup>19)</sup>. The error,  $\pm 1\%$ , of each count is taken into account in addition to the uncertainty,  $\pm 1\%$ , of the maximum electron energy.

#### § 3.1 The method of $W$ expansion

The  $\beta$ -spectrum of RaE has been precisely measured by many persons<sup>19,20)</sup>. The

experimental correction factor obtained by Plassman and Langer<sup>19)</sup> is shown in Fig. 1, in which we determined the width of the curve by taking account of the uncertainties,  $\pm 1\%$  in the maximum energy  $W_0$  and  $\pm 1\%$  in each count. In order to compare it with our effective radii theory (eqs. (6) to (8)), we expand the experimental curve into the finite power series in  $W$  as shown in eqs. (6). By tedious but straightforward calculations we can prove that the experimental curve in Fig. 1 leads to

$$-a_1/a_2 > 7, \text{ if } a_2 > 0, \quad (9)$$

which is implied in (8).

On the other hand, using eqs. (7) and (8) and the Fierz condition (2), we get the theoretical expression for  $a_1/a_2$  as :

$$\begin{aligned} a_1/a_2 = & \left[ \frac{4}{3} \left\{ \frac{\alpha Z}{2\rho_1(\beta \mathbf{r})} - \frac{2}{3} W_0 \right\} \varepsilon_s^2 \left| \int \beta \mathbf{r} I_1(r) \right|^2 \right. \\ & + \left\{ \frac{4}{3} \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} - \frac{5}{9} W_0 \right\} \varepsilon_A^2 \left| \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2 \\ & - \frac{4}{3} \alpha_{ST} \varepsilon_S \varepsilon_T (i \int \beta \mathbf{r} I_1(r)) * \left\{ \left( \int \beta \mathbf{a} I_1(r) \right) - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})} \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right) \right\} \\ & - \frac{4}{3} \alpha_{VA} \varepsilon_r \varepsilon_A (i \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r)) * \left\{ \left( \int \mathbf{a} I_1(r) \right) - i \frac{\alpha Z}{2\rho_1(\mathbf{r})} \left( \int \mathbf{r} I_1(r) \right) \right\} \\ & - \frac{1}{9} W_0 \varepsilon_T^2 \left| \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right| - \frac{4}{9} W_0 \varepsilon_V^2 \left| \int \mathbf{r} I_1(r) \right|^2 \Big] / \\ & \left[ \frac{8}{9} \varepsilon_s^2 \left| \int \beta \mathbf{r} I_1(r) \right| + \frac{1}{9} \varepsilon_T^2 \left| \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2 \right. \\ & \left. + \frac{4}{9} \varepsilon_V^2 \left| \int \mathbf{r} I_1(r) \right|^2 + \frac{5}{9} \varepsilon_A^2 \left| \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \right|^2 \right]. \end{aligned} \quad (10)$$

From (10) we can easily see that the interference terms between  $S$  and  $T$  and between  $V$  and  $A$  ( $\alpha_{ST}$  and  $\alpha_{VA}$  terms) play important parts to make  $-a_1/a_2$  large enough to fit the experimental data. In fact, if we put  $\alpha_{ST}=0$  and  $\alpha_{VA}=0$  in (10), we obtain the inequality relation,

$$-a_1/a_2 < W_0 = 3.29, \quad (11)$$

unless we take negative values for  $\rho_1(\beta \mathbf{r})$  and  $\rho_1(\boldsymbol{\sigma} \times \mathbf{r})$ .

Here, let us consider about these effective nuclear radii<sup>17)</sup>. An effective radius is defined as a ratio of two nuclear matrix elements whose operators have the same angular part but somewhat different radial dependences. We can divide one of the operators into two parts, one (c) has the same radial dependence as the other operator and another (d) has an entirely different radial dependence. Let us call them part c and part d respectively. The magnitude of the part d as an operator, not as a matrix element is about 1/25 of the part c in our case<sup>17)</sup>. If we neglect the part d we get an effective



radius of about 1.1 times the ordinary radius. An effective radius much different from this value takes place only when cancellation occurs in the radial integral of the part c and not in that of the part d. In order to get a negative effective radius, cancellation down to about  $1/25$  is necessary. That is to say in our case, the radial integral of every concerning nuclear matrix element,  $\int \beta \mathbf{r} I_1(r)$ ,  $[\alpha Z (2\rho_1(\beta \mathbf{r}))^{-1}] \int \beta \mathbf{r} I_1(r) \cdot \int \sigma \times \mathbf{r} I_1(r)$ , or  $[\alpha Z (2\rho_1(\sigma \times \mathbf{r}))^{-1}] \int \sigma \times \mathbf{r} I_1(r)$ , is required to take a value of about  $1/25$  of the possible maximum value. On the other hand the nuclear matrix elements  $\int \beta \mathbf{r} I_1(r)$  and  $\int \sigma \times \mathbf{r} I_1(r)$  are essential to make the absolute value of  $a$  large enough, as can be seen from (5) or (7). In order to produce a strongly energy dependent correction factor as that of RaE decay with (uncorrected)  $\log ft=8.0$ , either  $\int \beta \mathbf{r} I_1(r)$  or  $\int \sigma \times \mathbf{r} I_1(r)$  must be fairly large. Magnitude of a nuclear matrix element  $\int X$  can be expressed by a corrected  $ft$  value,  $f(X)t=2\pi^3 \ln 2 (\epsilon_f/\epsilon_i)^{1/2} \langle X \rangle$ , where  $\epsilon_i$  is the relevant coupling constant.  $f(X)t$  is inversely proportional to  $|\int X|^2$ . The magnitudes of  $\int \beta \mathbf{r} I_1(r)$  and  $\int \sigma \times \mathbf{r} I_1(r)$  necessary to explain the absolute energy dependence of the RaE spectrum correspond to  $\log f(\beta \mathbf{r} I_1(r))t \leq 9.4$  and  $\log f(\sigma \times \mathbf{r} I_1(r))t \leq 9.4$ .\*

If  $\log f(\beta \mathbf{r} I_1(r))t$  or  $\log f(\sigma \times \mathbf{r} I_1(r))t=9.6$ , we get a correction factor ( $\log ft=8.0$ ) whose lowest energy part is at most only 150% larger than the highest energy part. Now all the known  $\log ft$  values for the first forbidden transitions are not smaller than 5.2<sup>21)</sup>. The smallest values occur for  $\Delta J=0$  transitions. If we take account of the spin statistical factor,  $(2J_f+1)/(2J_i+1)$  ( $=1/3$  for RaE decay) or more conservatively  $[(2J_f+1)/(2J_i+1)]^{1/2}$ \*\*, the possible minimum  $\log ft$  value for  $J=1^- \rightarrow 0^-$  transitions is inferred to be about 5.5. It is very likely that this minimum value occurs only when there is no appreciable destructive interference in any respect. Thus we arrive at the conclusion that for a  $J=1^- \rightarrow 0^-$  transition the possible minimum values of  $\log f(\beta \mathbf{r} I_1(r))t$  and  $\log f(\sigma \times \mathbf{r} I_1(r))t$  are equal to  $5.5+2.3=7.8$ , where the  $2.3=\log [\alpha Z/(2\rho)]^2$  came from the Coulomb factor. Comparing this 7.8 with 9.4 which we obtained as the possible maximum value for RaE decay, we see that the radial integrals of  $\int \beta \mathbf{r} I_1(r)$  and  $\int \sigma \times \mathbf{r} I_1(r)$  take the value of at least  $10^{(9.4-7.8)/2} \approx 1.6$  times the possible maximum value. This result excludes the possibility of negative effective radius, for which the value of  $>1/25$  is necessary instead of 1/6. Thus the variations of  $\rho_1(\beta \mathbf{r})$  and  $\rho_1(\sigma \times \mathbf{r})$  are confined to

\* The rough evaluation of these upper bound can be made by using the approximate form of the correction factor,

$$\xi^2 + \alpha \frac{4}{3} W |\xi| + \frac{4}{9} W^2 |x|^2,$$

where  $\xi$  and  $\alpha$  are energy independent and  $x$  is  $\int \beta \mathbf{r} I_1(r)$  or  $\int \sigma \times \mathbf{r} I_1(r)$ . The corresponding  $f(x)$  values is

$$f(X) = \int F_0(W, Z) \left[ \xi^2 + \alpha \frac{4}{3} W \xi + \frac{4}{9} W^2 \right] \rho W q^2 dW.$$

The upper bound given in the text is taken somewhat larger than this rough estimate making allowance for the incorrectness of the above formulas.

\*\* We cannot define the general statistical factor uniquely. The former example in the text is only the ratio of the number of final substates of the decay in question to that of a hypothetical  $\Delta J=0$  transition starting from the same nucleus. The latter is suggested by the fact that the reduced transition probability is  $(2J_f+1)/(2J_i+1)$  times that of the reverse process.

$$(1-6/25)1.1\rho > \rho_1(\beta\mathbf{r}), \quad \rho_1(\boldsymbol{\sigma} \times \mathbf{r}) < 1.1\rho/(1-6/25),$$

i.e.,

$$0.8\rho < \rho_1(\beta\mathbf{r}), \quad \rho_1(\boldsymbol{\sigma} \times \mathbf{r}) < 1.5\rho. \quad (12a)$$

For the later purpose (see § 4) let us consider the case where we take 9.6 as the possible maximum value of  $\log f(\beta\mathbf{r}I_1(r))t$  and  $\log f(\boldsymbol{\sigma} \times \mathbf{r}I_1(r))t$  for RaE decay. In this case the radial integrals of  $\int \beta\mathbf{r}I_1(r)$  and  $\int \boldsymbol{\sigma} \times \mathbf{r}I_1(r)$  take the values of at least  $1/8$  times the possible maximum value. The variations of  $\rho_i$ 's are

$$0.7\rho > \rho_1(\beta\mathbf{r}), \quad \rho_1(\boldsymbol{\sigma} \times \mathbf{r}) < 1.7\rho. \quad (12b)$$

Thus we were able to establish (11) on solid foundation.

The relation (11) clearly contradicts with the experimental fact (9). Thus we can reasonably exclude the case of

$$\alpha_{ST}=0 \text{ and } \alpha_{VA}=0. \quad (13)$$

It is interesting to note that the above relation holds for the following special cases:

- a) Only one type of  $S$ ,  $V$ ,  $T$  or  $A$ .
- b)  $VT$  type.
- c)  $SA$  type.
- d)  $ST$  type with  $C_S/C_T = C'_S/C'_T = \text{pure imaginary}$ .
- e)  $VA$  type with  $C_V/C_A = C'_V/C'_A = \text{pure imaginary}$ .
- f)  $ST$  type with  $C_S/C'_S = -C'_T/C_T = \text{real}$ .
- g)  $VA$  type with  $C_V/C'_V = -C'_A/C_A = \text{real}$ .

In the cases d) and e) the two component neutrino theory<sup>10)</sup> is valid, but the time reversal invariance<sup>7)</sup> is not. In the cases f) and g) the two component theory is not valid. In the next two subsections we investigate the special cases  $ST$  and  $VA$  in detail.

### § 3.2 $ST$ type

In this section we examine a possible special case,  $ST$  type, suggested by the data on electron-neutrino angular correlation of  $\text{He}^{622}$ . In this case (5) can be written as

$$\begin{aligned} C_{1ST}(W) = \varepsilon_T^2 \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r) \left[ X^2 + \left\{ \left( \frac{\alpha Z}{2\rho_1(\beta\mathbf{r})} \right)^2 - \frac{2}{3} \left( q - \frac{p^2}{W} \right) \frac{\alpha Z}{2\rho_1(\beta\mathbf{r})} \right. \right. \\ \left. \left. + \frac{q^2 + p^2}{3} - \frac{2qp^2}{9W} \right\} Y^2 + \left\{ \frac{2}{3} \left( q - \frac{p^2}{W} \right) - 2 \frac{\alpha Z}{2\rho_1(\beta\mathbf{r})} \right\} \alpha_{ST} XY \right. \\ \left. - \frac{2}{3} \left( q + \frac{p^2}{W} \right) X + \frac{2}{3} \left( q + \frac{p^2}{W} \right) \frac{\alpha Z}{2\rho_1(\beta\mathbf{r})} \alpha_{ST} Y + \frac{q^2 + p^2}{6} + \frac{2qp^2}{9W} \right], \quad (14) \end{aligned}$$

where

$$X = \frac{\int \beta a I_1(r)}{\int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r)} - \frac{\alpha Z}{2\rho_1(\beta \boldsymbol{\sigma} \times \mathbf{r})}, \quad Y = \frac{i\varepsilon_S \int \beta \mathbf{r} I_1(r)}{\varepsilon_T \int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r)}. \quad (15)$$

According to the usual method for analysing  $\beta$ -spectra, we choose the three electron energies,  $W_1=1.72$ ,  $W_2=2.25$ ,  $W_3=2.75$ , arbitrarily. The requirement that the theoretical correction factor (14) runs through the experimental region of  $C(W)$  on Fig. 1 at  $W=W_1$  and  $W_2$  (or  $W=W_1$  and  $W_3$ ) restricts the allowed region of the parameters,  $X$  and  $Y$ . In the actual analysis we normalize  $C(W)$  at  $W=1.72$  according to Plassman and Langer<sup>19</sup>). We obtain the ratios

$$C(W_1)/C(W_2)=1.40 \text{ to } 1.50, \quad (16a)$$

$$C(W_1)/C(W_3)=1.96 \text{ to } 2.32, \quad (16b)$$

taking into account the experimental errors. The allowed region of the parameters,  $X$  and  $Y$ , lies between the four curves given by eqs. (16a) and (16b) with the limiting values in the right-hand sides.

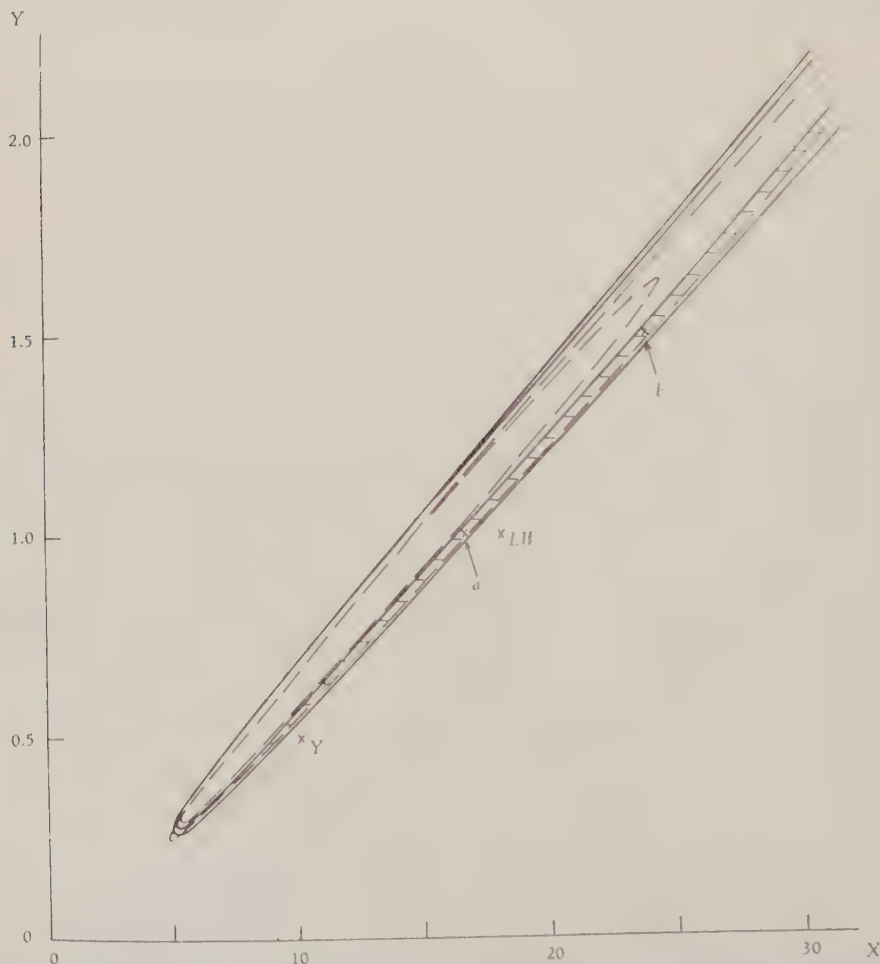


Fig. 2. The shaded area is the allowed region for the parameters,  $X$  and  $Y$ , in the  $ST$  case with  $\alpha_{ST}=1$ .  $Y$  and  $LW$  are the points used by Yamada<sup>2)</sup> and Lee-Whiting respectively. For  $a$  and  $b$  the correction factors are shown on Fig. 3.



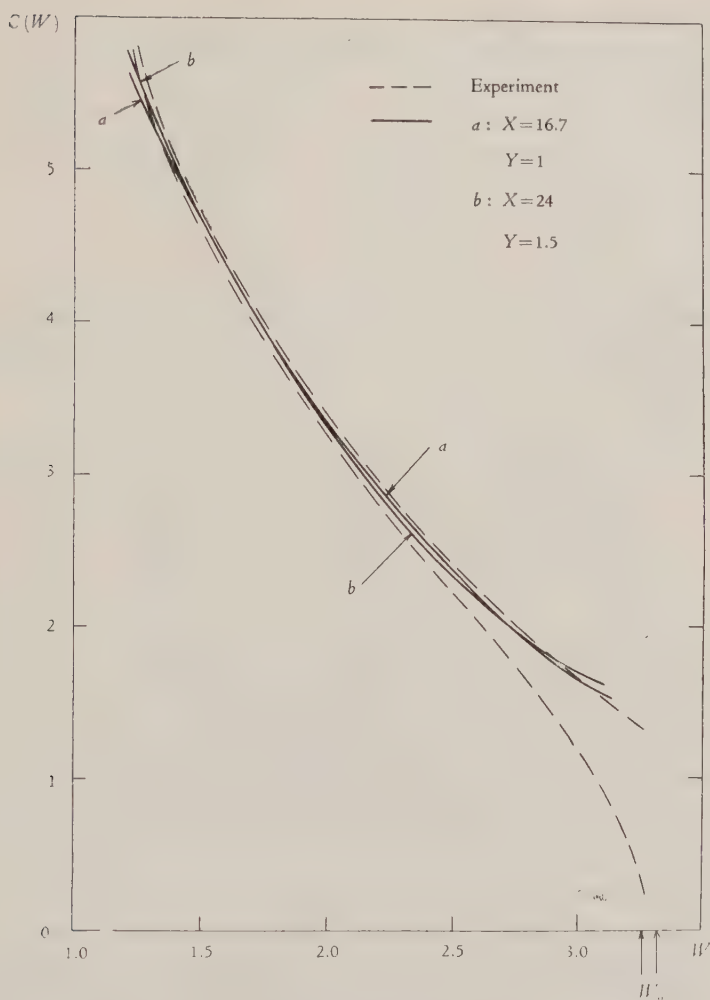


Fig. 3. Correction factors for  $ST$  with  $\alpha_{ST}=1$ .

In order to compare the effective nuclear radii theory with the previous analyses, we first treat the case,  $\alpha_{ST}=1$ . For simplicity, we adopt a fixed value,  $W_0=3.29$ , for the maximum electron energy in the theoretical expression (14), because the uncertainty is already taken into account in Fig. 1. Furthermore we put  $\alpha Z/[2\rho_1(\beta\mathbf{r})]=13.25$ , in which the effective nuclear radius  $\rho_1(\beta\mathbf{r})$  is assumed to be 25% larger than the ordinary nuclear radius,  $\rho=1.2A^{1/3}\times 10^{-13}$  cm. The region of the parameters thus obtained is shown as the narrow shaded area in Fig. 2. The previous parameters used by Yamada<sup>2)</sup> and Lee-Whiting<sup>2)</sup> are plotted as the crosses in the figure. Our allowed region may be shifted to include these crosses by choosing a somewhat different value for  $\rho_1(\beta\mathbf{r})$ . To visualize our results we plot in Fig. 3 the correction factor for special choices of the parameters,  $X=16.7$  and  $Y=1$ , and  $X=24$  and  $Y=1.5$ . It should be noted that the

above results are unchanged for  $\alpha_{ST} = -1$  if we replace  $Y$  by  $-Y$ . Therefore the sign of  $\alpha_{ST}$  cannot be determined from the present analysis unless we know the relative signs of the concerning nuclear matrix elements.

Now let us examine the case,  $|\alpha_{ST}| < 1$ . In this case it is more difficult to explain the experimental shape than in the case,  $|\alpha_{ST}| = 1$ . The restrictions for  $|\alpha_{ST}|$  are summarized in Table I for a few assumed values of  $\rho_1(\rho)$  which cover the allowable variation discussed in the previous subsection.

Table I. Restrictions for degrees of interferences between  $S$  and  $T$ ,  $\alpha_{ST}$ , and between  $V$  and  $A$ ,  $\alpha_{VA}$ , which are obtained from analyses of RaE spectrum with the effective nuclear radii theory. Only  $\rho_1, \rho$  values satisfying (12a) can actually occur.

type Eq. used	ST		VA	
	(16a)	(16b)	(16a)	(16b)
$\rho_1/\rho$	$ \alpha_{ST}  >$	$ \alpha_{ST}  >$	$ \alpha_{VA}  >$	$ \alpha_{VA}  >$
0.4	0.9991	0.9993	0.9995	0.9996
0.6	0.9980	0.9985	0.9988	0.9990
1.25	0.992	0.994	0.995	0.996
2	0.982	0.987	0.989	0.991
3	0.966	0.975	0.979	0.983

For the excluded values of  $|\alpha_{ST}|$ , at least one of the left-hand sides of (16a) and (16b) is always smaller than the right-hand side. The allowed deviations of  $\alpha_{ST}$  from unity are quite small irrespective of the values of the effective nuclear radius, suggesting the actual occurrence of the maximum interference,  $|\alpha_{ST}| = 1$ .

### § 3.3 VA type

Another possible type besides  $ST$  is  $VA$  as suggested by the recent experiments on electron-neutrino angular correlation of  $A^{35\text{Co}}$  and on circular polarization of  $\gamma$ -rays following the electron capture of  $\text{Eu}^{152\text{Eu}}$ . The expression of the correction factor for  $VA$  can be obtained only by reversing the sign of the neutrino momentum  $q$  in the correction factor for  $ST$  in § 3—2, hence they are quite similar. Defining the parameters,  $X$  and  $Y$ , as

$$\begin{aligned}
 X &= \frac{\int \mathbf{r} I_1(r)}{i \int \mathbf{r} I_1(r)} - \frac{\alpha Z}{2\rho_1(\mathbf{r})}, \\
 Y &= \frac{\varepsilon_A \int \boldsymbol{\sigma} \times \mathbf{r} I_1(r)}{i \varepsilon_V \int \mathbf{r} I_1(r)},
 \end{aligned} \tag{17}$$

we get the following theoretical correction factor:

$$C_{VA}(W) = \varepsilon_V^2 \left| \int \mathbf{r} I_1(r) \right|^2 \left[ X^2 + \left\{ \left( \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} \right)^2 - \frac{2}{3} \left( q - \frac{p^2}{W} \right) - \frac{\alpha Z}{2\rho_1(\boldsymbol{\sigma} \times \mathbf{r})} \right\} \right]$$

$$\begin{aligned}
 & + \frac{q^2 + p^2}{6} - \frac{2qp^2}{9W} \} Y^2 + \left\{ \frac{2}{3} \left( q - \frac{p^2}{W} \right) - 2 \frac{\alpha Z}{2\rho_1(\sigma \times \mathbf{r})} \right\} \alpha_{VA} XY \\
 & - \frac{2}{3} \left( q + \frac{p^2}{W} \right) X + \frac{2}{3} \left( q + \frac{p^2}{W} \right) \frac{\alpha Z}{2\rho_1(\sigma \times \mathbf{r})} \alpha_{VA} Y + \frac{q^2 + p^2}{3} + \frac{2qp^2}{9W} \Big] \quad (18)
 \end{aligned}$$

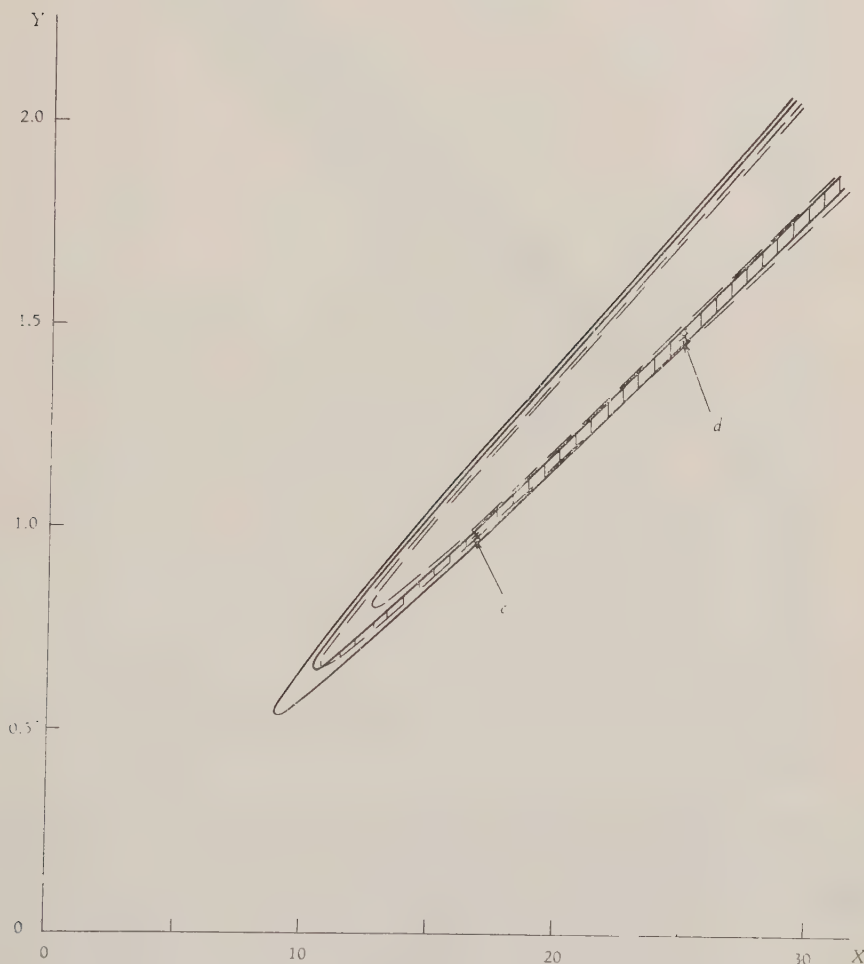


Fig. 4. The shaded area is a part of the allowed region for the parameters,  $X$  and  $Y$ , in the  $VA$  case with  $\alpha_{VA}=1$ . For  $c$  and  $d$  the correction factors are shown on Fig. 6.

Analyses were made in the same way as the  $ST$  case. First, for  $\alpha_{VA}=1$  allowed regions for the parameters,  $X$  and  $Y$ , are shown in Figs. (4) and (5). The only main difference in this case is that the allowed parameter regions exist for both signs of  $Y$  contrary to the  $ST$  case, two typical correction factors are shown in Fig. 6. The fit is as good as that for the  $ST$  type. For  $\alpha_{VA}=-1$  the results are the same if we reverse the sign of  $Y$ .

Then we calculate the case of  $|\alpha_{VA}| < 1$ . The allowed regions for  $|\alpha_{VA}|$  are sum-



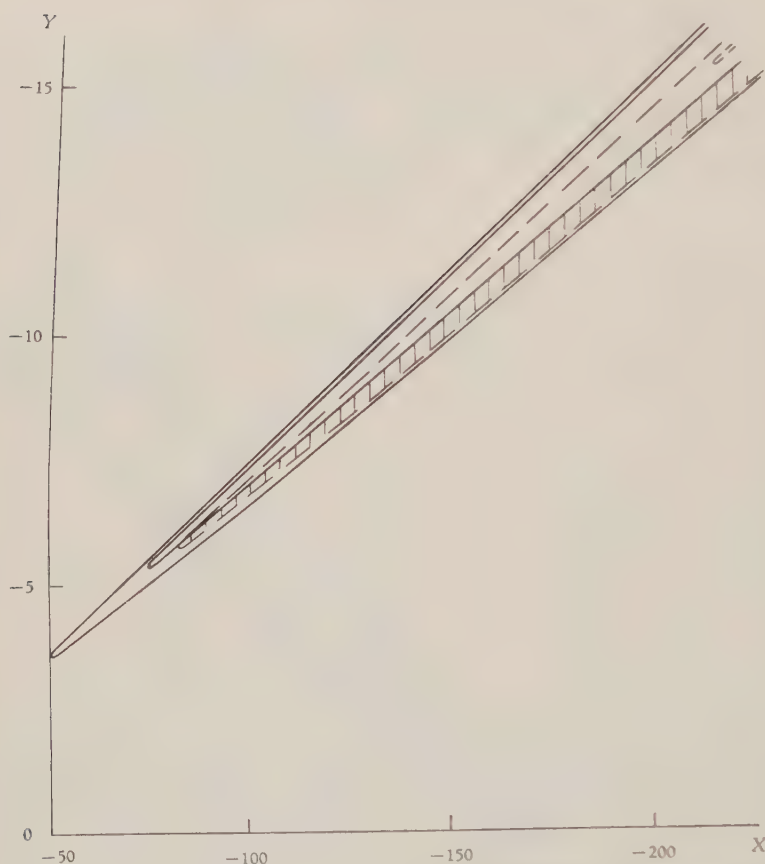


Fig. 5. The shaded area is a part of the allowed region for the parameters,  $X$  and  $Y$ , in the  $VA$  case with  $\alpha_{VA}=1$ .

marized in Table I for several assumed values of  $\mu_i(\sigma \setminus \mathbf{r})$ ,  $\mu$  which cover the allowable variation of the effective radius. The smallness of the allowed deviations of  $\alpha_{r_i}$  from unity suggests the actual occurrence of  $|\alpha_{r_i}|=1$ .

### § 34. SVTA type

The case of SVTA mixing is qualitatively surveyed in this subsection. It can be inferred from the argument of § 3.1 that the RaE spectrum favors  $\alpha_{r_i} = \alpha_{r_{i+1}} = 1$ . In such a case all the Fierz terms (terms having 1  $H^*$  factor in (5)) vanish, and the correction factor can be written as:

$$C_{1SVTA}(W) = C_{1ST}(W) + C_{1VA}(W). \quad (19)$$

Therefore, in the case of SVTA mixing, independent cancellations must occur simultaneously in both parts of  $ST$  and  $VA$  in order to explain the RaE spectrum. Although we cannot exclude this possibility definitely, the occurrence of such an event is expected

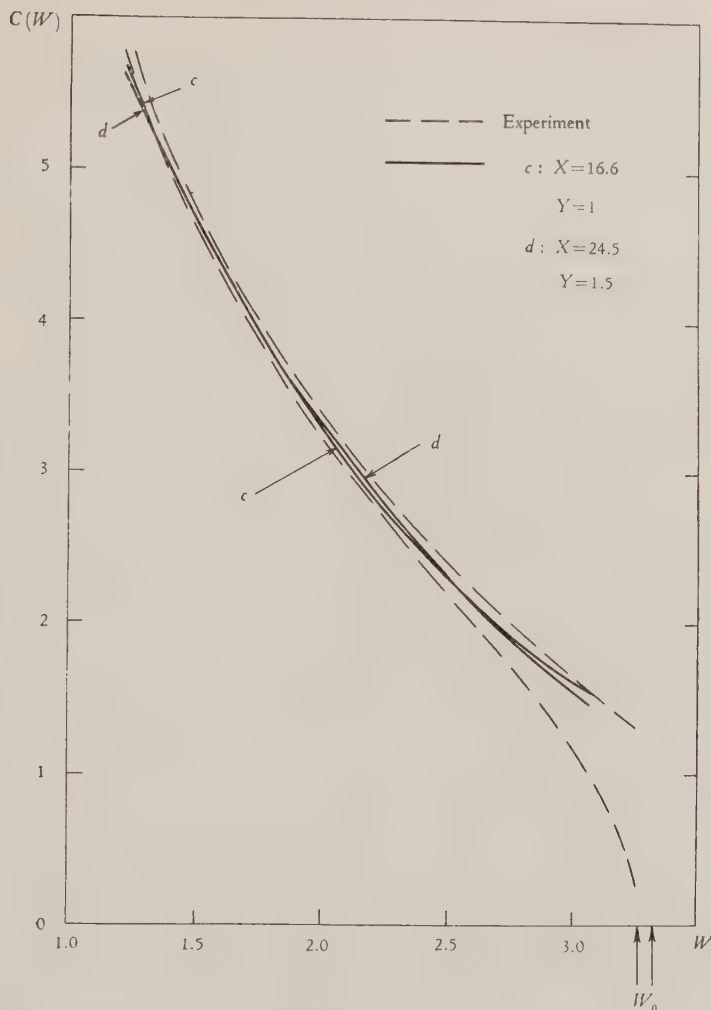


Fig. 6. Correction factors for  $VA$  with  $\alpha_{VA}=1$ .

to be highly accidental. Hence, the RaE spectrum favors either  $ST$  or  $VA$ , but not  $SVTA$  mixing, from the view point of probabilism.

It can be shown from analyses of Figs. 2,4 and 5 that if the relation,

$$\varepsilon_S \varepsilon_A / \varepsilon_V \varepsilon_T = YY' > 0.16,$$

is satisfied for the case of  $|\alpha_{ST}| = |\alpha_{VA}| = 1$  there certainly exist suitable parameters to explain the experimental correction factor.

#### § 4. Evaluation of the effects of neglected terms

Although we obtained very stringent restrictions for  $|\alpha_{ST}|$  and  $|\alpha_{VA}|$  in § 3, the arguments were based on the validity of the effective radii theory. This theory seems to be sufficiently accurate for usual analyses, but in order to deduce definitive conclusions

we must examine the terms neglected in that theory.

Aside from the radiative correction, three approximations are made in the effective radii theory: 1) approximation to the inner wave functions of electron, 2) approximation to the normalization of the wave functions of electron, and 3) restriction to the number of partial waves to be taken into account. The effect of the approximation 2) does not seem to be significant, because no new nuclear matrix element is concerned and the effect is not magnified even when there is a large cancellation<sup>17</sup>. On the other hand new nuclear matrix elements appear if we eliminate the approximations 1) and 3), and their effects are magnified when cancellations occur among the main terms. Therefore, we evaluate the effect of the approximations 1) and 3) in this section.

If we denote the correction factor  $C_{1ST}(W)$  or  $C_{1T1}(W)$  in the effective radii theory  $C_1(W)$ , the correction factor that includes the above mentioned corrections are approximately given by the following expression:

$$C(W) \approx C_1(W) \pm 2|G(W)|\sqrt{C_1(W)} + |G(W)|^2, \quad (20)$$

where the  $G(W)$  is

$$\begin{aligned} G_{ST}(W) = & -\frac{W^2+1}{W} \varepsilon_T \left( \int_0^r \beta a r^{-1} dr_1 r_1^2 V(r_1) I_1(r_1) \right) \\ & + \frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} \varepsilon_T \left( \int \beta a K_1^{(2)}(r) \right) \\ & - \frac{q}{3} \frac{\alpha Z}{2\rho_1(\beta a r^2)} \varepsilon_T \left( \int \beta a r^2 I_1(r) \right) \\ & - \frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} \varepsilon_T \left( \int \beta \boldsymbol{\sigma} \times \mathbf{r} K_1^{(1)}(r) \right) \\ & - \frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} i \alpha_{ST} \varepsilon_S \left( \int \beta \mathbf{r} K_1^{(1)}(r) \right) \\ & + \frac{2q}{3} \frac{\alpha Z}{2\rho_1(\mathbf{r}(\beta \mathbf{a} \cdot \mathbf{r}))} \varepsilon_T \left( \int \mathbf{r} (\beta \mathbf{a} \cdot \mathbf{r}) I_1(r) \right), \end{aligned} \quad (21a)$$

for  $ST$  and

$$\begin{aligned} G_{VA}(W) = & -\frac{W^2+1}{W} \varepsilon_V \left( \int a r^{-1} dr_1 r_1^2 V(r_1) I_1(r_1) \right) \\ & + \frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} \varepsilon_V \left( \int a K_1^{(2)}(r) \right) \\ & + \frac{q}{3} \frac{\alpha Z}{2\rho_1(a r^2)} \varepsilon_V \left( \int a r^2 I_1(r) \right) \\ & - \frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} \alpha_{VA} \varepsilon_A \left( \int \boldsymbol{\sigma} \times \mathbf{r} K_1^{(1)}(r) \right) \end{aligned}$$



$$\begin{aligned}
 & -\frac{2(2W^2+1)}{3W} \frac{\gamma_1}{D_{01}} i\varepsilon_V \left( \int \mathbf{r} K_1^{(1)}(r) \right) \\
 & -\frac{2q}{3} \frac{\alpha Z}{2\rho_1(\mathbf{r}(\alpha \cdot \mathbf{r}))} \varepsilon_T \left( \int \mathbf{r}(\alpha \cdot \mathbf{r}) I_1(r) \right), \quad (21b)
 \end{aligned}$$

for  $VA$ .  $K_1^{(1)}(r)$  and  $K_1^{(2)}(r)$  are defined in eqs. (3) of ref. 17),  $D_{01}$  in eq. (12) of ref. 17) and  $\gamma_1 = \sqrt{1 - \alpha^2 Z^2}$ . In (20) the second term in the right-hand side is the main correction term, and shows strong energy dependence. Its form, given by (20) and (21a) or by (20) and (21b), is approximately the upper bound (in magnitude) of the exact and more complicated form. The last term in (20) is generally small, and has a negative effect to produce strong energy dependence as is required for RaE decay. Its form given by (20) and (21a), or by (20) and (21b), is the lower bound of the exact form. Thus in the approximate form (20) with (21a) or (21b) the energy dependence due to the correction terms is fully taken into account.

In order to see how much effect the correction terms could have, we first consider the upper bounds of the new nuclear matrix elements. Generally it seems reasonable to set the upper bound of the nuclear matrix element  $\int \gamma U_1(r)$ , where  $\gamma$  is independent of  $r$ , as  $\int_0^p r^2 U_1(r) dr / \int_0^p r^2 U_2(r) dr$  times the upper bound of  $\int \gamma U_2(r)$ . Using this relation and assuming the uniform charge distribution for the nucleus, we can relate the upper bounds of the nuclear matrix elements, except  $\int \mathbf{r}(\beta \alpha \cdot \mathbf{r}) I_1(r)$  and  $\int \mathbf{r}(\alpha \cdot \mathbf{r}) I_1(r)$ , with the familiar first forbidden matrix elements. We do not know much about the new matrix elements,  $\int \mathbf{r}(\beta \alpha \cdot \mathbf{r}) I_1(r)$  and  $\int \mathbf{r}(\alpha \cdot \mathbf{r}) I_1(r)$ . We assume that their upper bounds are equal to those of  $\int \beta \alpha^2 I_1(r)$  and  $\int \alpha^2 I_1(r)$  respectively. According to the argument of § 3.1 we can set the upper bounds of  $\int \alpha I_1(r)$ ,  $\int \beta \alpha I_1(r)$ ,  $\int \mathbf{r} I_1(r)$ ,  $\int \beta \mathbf{r} I_1(r)$ ,  $\int \boldsymbol{\sigma} \times \mathbf{r} I_1(r)$  and  $\int \beta \boldsymbol{\sigma} \times \mathbf{r} I_1(r)$  as those corresponding to the corrected  $\log ft$  values ( $\log f(\alpha I_1(r))t$  etc., (see § 3.1) of 5.5, 5.5, 7.8, 7.8, 7.8 and 7.8 respectively. We take these upper bounds of the nuclear matrix elements and give them appropriate signs and put  $|\alpha_{ST}| = |\alpha_{VA}| = 1$  so as to make  $|G_{ST}(W)|$  and  $|G_{VA}(W)|$  maximum. In this procedure the relative signs of the first three terms in the right-hand sides of (21a) and (21b) were not adjusted because they are similar and not independent of each other. Using those  $|G(W)|$ 's and the experimental  $C(W)$  (including normalization), we can calculate  $C_1(W)$  from (20). This  $C_1(W)$  has the possible least energy dependence.  $C_1(W)$ 's thus obtained for  $ST$  and  $VA$  are shown in Fig. 7. To obtain them only the  $C(W)$  with smallest energy dependence allowed by Fig. 1 was used. We can see in Fig. 7 that the correction factor of the lowest energy part is still more than 150 % larger than that of the highest energy part. This fact was already taken into account in estimating the flexibility of the effective radii in § 3.1. From Fig. 7 we get

$$C_1(W_1)/C_1(W_2) = \begin{cases} 1.26 & \text{for } ST, \\ 1.22 & \text{for } VA, \end{cases} \quad (22a)$$

and

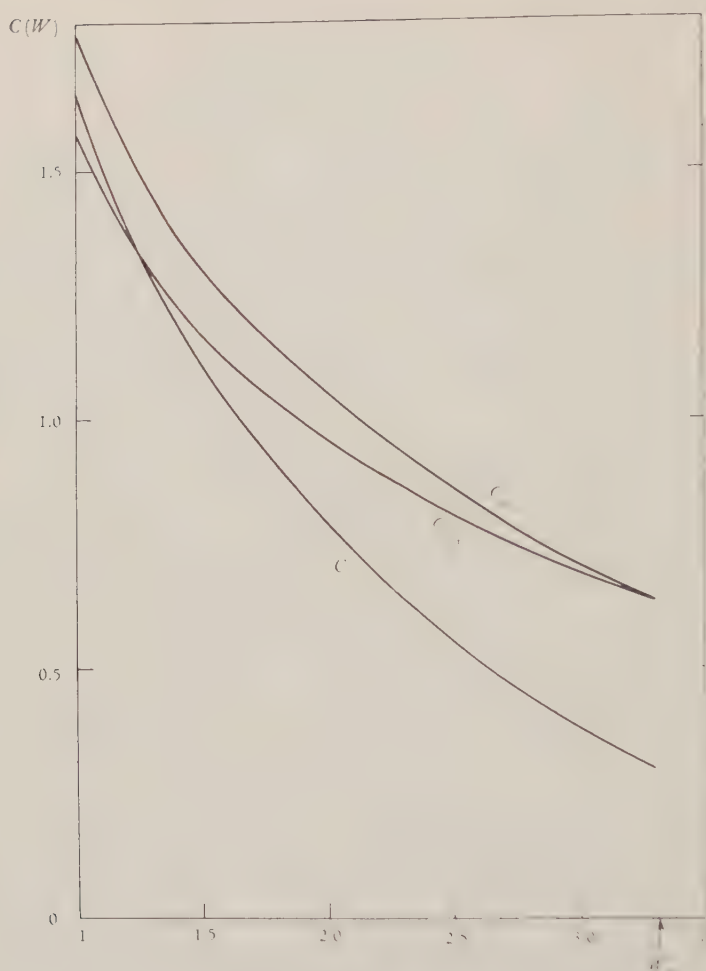


Fig. 7. Corrected correction factors  $C_1(W)$  which are obtained by subtracting possible maximum effects of (theoretical) correction terms from the experimental one. For detail see § 4.

$$C_1(W_1)/C_1(W_3) = \begin{cases} 1.54 & \text{for } ST, \\ 1.45 & \text{for } VA. \end{cases} \quad (22b)$$

The right-hand sides of (22a) and (22b) are valid only as the lower limits.

Using (22a) and (22b) we can calculate in the same way as in § 3.2 and § 3.3. The limitations for  $|\alpha_{ST}|$  and  $|\alpha_{VA}|$  are summarized in table II for several values of  $\rho_1/\rho$  which cover possible variation of  $\rho_1$ , (12b). The restrictions are somewhat weakened from those in Table I. However, the interference terms must be still about maximum.

Although we made somewhat doubtful assumptions in estimating the upper bounds of the nuclear matrix elements in the correction terms, it will be well compensated by the assumption that all the matrix elements take the possible maximum values simul-

Table II. Restrictions for degrees of interferences between  $S$  and  $T$ ,  $|\alpha_{ST}|$ , and between  $V$  and  $A$ ,  $|\alpha_{VA}|$ , which are obtained from analyses of RaE spectrum with the effective nuclear radii theory including possible maximum effect of correction terms. Only  $\rho_1/\rho$  values satisfying (12b) can actually occur.

type	ST		VA	
Eq. used	(22a)	(22b)	(22a)	(22b)
$\rho_1/\rho$	$ \alpha_{ST} >$	$ \alpha_{ST} >$	$ \alpha_{VA} >$	$ \alpha_{VA} >$
0.4	0.997	0.998	0.998	0.998
0.6	0.995	0.995	0.996	0.996
1.25	0.982	0.985	0.987	0.987
2	0.961	0.968	0.973	0.974
3	0.911	0.942	0.950	0.950

taneously and are added constructively. Suppose that if this occurs for the ordinary first forbidden matrix elements the  $\log ft$  is about 4.2 for a  $J \rightarrow J \neq 0$  transition, which corresponds to a fairly fast allowed transition. Thus we can regard the values in Table II as giving safe limits of interference terms.

## § 5. Discussions and conclusions

In this paper we have assumed the following points:

- The experimental correction factor for the  $\beta$ -spectrum of RaE which was measured by Plassman and Langer<sup>19)</sup>.
- Validity of the generalized Fermi theory for  $\beta$ -decay, in which neither derivative nor non-local interaction is included.
- Applicability of the "effective nuclear radii theory"<sup>217)</sup> with some correction terms.
- Validity of the time reversal invariance of the Wigner type for the strong nuclear interactions.

We shall discuss the validity of these assumptions in the following.

Our results will not be changed significantly even if we use other experimental data. Almost the same spectrum were obtained by the independent experiment of Wu and Lidofsky<sup>20)</sup>.

We have no definite evidence that any theory of  $\beta$ -decay other than the Fermi theory is successful. The derivative interactions as proposed by Konopinski and Uhlenbeck<sup>5)</sup> might be successful for explaining the RaE spectrum, but contradict with  $\beta$ -spectra of most of other nuclei.

In connection with the assumption c), we should mention that the radiative correction<sup>25)</sup> is omitted in this paper. Although this correction seems to be negligible from both the theoretical arguments and the absence of positive experimental proof of its existence in any  $\beta$ -decay, we cannot make definite statement about it. Also we cannot be completely sure that our estimation of the upper bounds of the nuclear matrix elements are all correct, though the failure of our argument about it seems to be highly im-



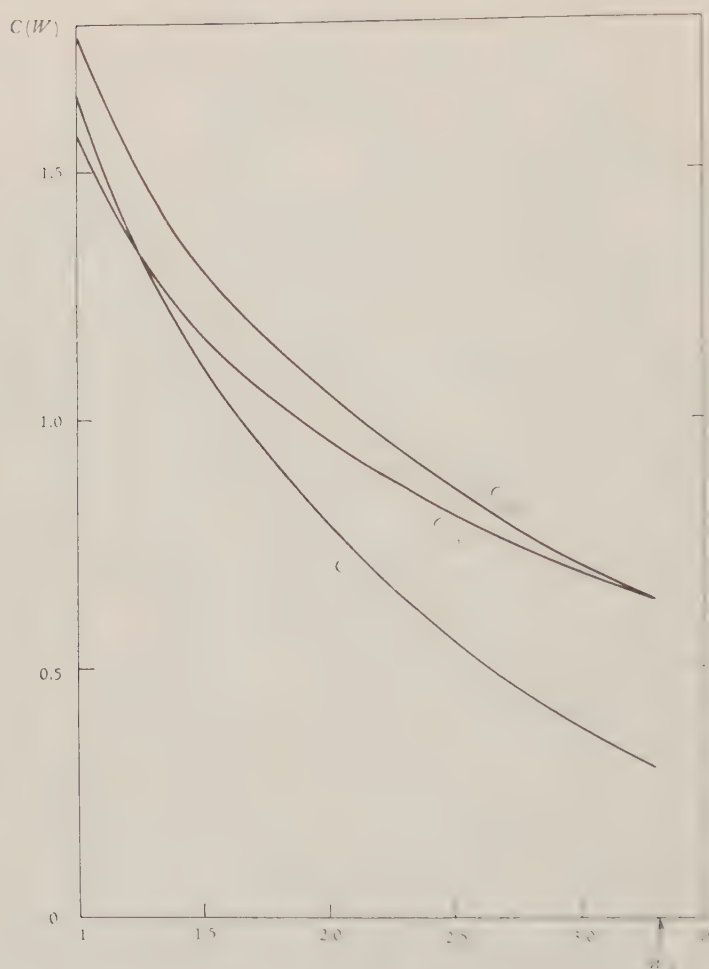


Fig. 7. Corrected correction factors  $C_1(W)$  which are obtained by subtracting possible maximum effects of (theoretical) correction terms from the experimental one. For detail see § 4.

$$C_1(W_1)/C_1(W_3) = \begin{cases} 1.54 & \text{for } ST, \\ 1.45 & \text{for } VA. \end{cases} \quad (22b)$$

The right-hand sides of (22a) and (22b) are valid only as the lower limits.

Using (22a) and (22b) we can calculate in the same way as in § 3.2 and § 3.3. The limitations for  $|\alpha_{SP}|$  and  $|\alpha_{VA}|$  are summarized in table II for several values of  $\rho_1/\rho$  which cover possible variation of  $\rho_1$ , (12b). The restrictions are somewhat weakened from those in Table I. However, the interference terms must be still about maximum.

Although we made somewhat doubtful assumptions in estimating the upper bounds of the nuclear matrix elements in the correction terms, it will be well compensated by the assumption that all the matrix elements take the possible maximum values simul-

Table II. Restrictions for degrees of interferences between  $S$  and  $T$ ,  $|\alpha_{ST}|$ , and between  $V$  and  $A$ ,  $|\alpha_{VA}|$ , which are obtained from analyses of RaE spectrum with the effective nuclear radii theory including possible maximum effect of correction terms. Only  $\rho_1/\rho$  values satisfying (12b) can actually occur.

type	ST		VA	
Eq. used	(22a)	(22b)	(22a)	(22b)
$\rho_1/\rho$	$ \alpha_{ST}  >$	$ \alpha_{ST}  >$	$ \alpha_{VA}  >$	$ \alpha_{VA}  >$
0.4	0.997	0.998	0.998	0.998
0.6	0.995	0.995	0.996	0.996
1.25	0.982	0.985	0.987	0.987
2	0.961	0.968	0.973	0.974
3	0.911	0.942	0.950	0.950

taneously and are added constructively. Suppose that if this occurs for the ordinary first forbidden matrix elements the  $\log ft$  is about 4.2 for a  $J \rightarrow J \neq 0$  transition, which corresponds to a fairly fast allowed transition. Thus we can regard the values in Table II as giving safe limits of interference terms.

## § 5. Discussions and conclusions

In this paper we have assumed the following points:

- The experimental correction factor for the  $\beta$ -spectrum of RaE which was measured by Plassman and Langer<sup>19)</sup>.
- Validity of the generalized Fermi theory for  $\beta$ -decay, in which neither derivative nor non-local interaction is included.
- Applicability of the "effective nuclear radii theory"<sup>217)</sup> with some correction terms.
- Validity of the time reversal invariance of the Wigner type for the strong nuclear interactions.

We shall discuss the validity of these assumptions in the following.

Our results will not be changed significantly even if we use other experimental data. Almost the same spectrum were obtained by the independent experiment of Wu and Lidofsky<sup>20)</sup>.

We have no definite evidence that any theory of  $\beta$ -decay other than the Fermi theory is successful. The derivative interactions as proposed by Konopinski and Uhlenbeck<sup>5)</sup> might be successful for explaining the RaE spectrum, but contradict with  $\beta$ -spectra of most of other nuclei.

In connection with the assumption c), we should mention that the radiative correction<sup>25)</sup> is omitted in this paper. Although this correction seems to be negligible from both the theoretical arguments and the absence of positive experimental proof of its existence in any  $\beta$ -decay, we cannot make definite statement about it. Also we cannot be completely sure that our estimation of the upper bounds of the nuclear matrix elements are all correct, though the failure of our argument about it seems to be highly im-

probable. It should be noted that the mesic corrections<sup>2</sup> can be regarded as being included in our theory, because the nuclear matrix elements are treated as adjustable parameters and their upper bounds are determined on the empirical basis.

From the assumptions a), b), c) and d) we are led to the following conclusions:

- a) In the case of SVTA mixing,  $\alpha_{ST} = \alpha_{TA} = 0$  can be definitely excluded (see § 3.1).
- b) For ST mixing  $0.97_5 < |\alpha_{ST}| \leq 1$  (see Table II).
- c) For VA mixing  $0.97_9 < |\alpha_{TA}| \leq 1$  (see Table II).
- d) SVTA mixing seems to be unfavored from the view point of probabilism, although the RaE spectrum can be explained in principle if  $\alpha_{ST} \approx 1$  and  $\alpha_{TA} \approx 1$  (see § 3.4). These conclusions are consistent with most of other recent data<sup>17</sup> on the interference terms. Among all, our conditions are the most stringent and strongly suggest the actual occurrence of  $|\alpha_{ST}| = 1$  or  $|\alpha_{TA}| = 1$ . The equations  $\alpha_{ST} = 1$  and  $\alpha_{TA} = 1$  are equivalent to eqs. (3), from which follow the validities of the two component neutrino theory<sup>1</sup> and the time reversal invariance<sup>7)</sup> in  $\beta$ -decays.

Similar results were obtained by Yoshizawa et al.<sup>18</sup> with similar analyses on Cs<sup>137</sup> decay.

The authors express their sincere thanks to Dr. M. Morita and Dr. T. Kotani for repeated correspondences. They are also indebted to Dr. H. Takebe for valuable discussions, who has investigated the RaE spectrum using his theory<sup>2)</sup> (29). One of the authors (MY) thanks Dr. K. Way for her kind hospitality.

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## Mesic Correction to the Beta-Decay in a Nucleus

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In this paper the mesic corrections to the beta-decay interactions in a nucleus are investigated. It is found that the Gamow-Teller coupling constant should apparently become about 8% smaller by exchanging pions among nucleons. Also the mesic effect in the *l*-forbidden case is discussed.

## § 1. Introduction

It is known<sup>1)</sup> that the strong interaction between a nucleon and pions somewhat changes the type of the beta-decay interaction of the nucleon. Although for the beta-decay of a bare nucleon Gamow-Teller and Fermi coupling constants have just the same magnitude, that is,  $|g_{GT}^0/g_F^0|=1$ , it is expected that it deviates from unity owing to the mesic corrections. In fact, this theoretical expectation,<sup>2</sup> the apparent increase of the ratio, seems to be consistent with the empirical fact,<sup>3</sup> that  $|g_{GT}/g_F|=1.37$ , though the extent of the agreement is not clear, because the theory involves some ambiguities.

However, all the previous investigations<sup>1-3</sup> are based on the assumption that the nucleon can be regarded as a free particle. Actual nucleon exists in a nucleus, and the meson cloud should be a little suppressed by the exclusion principle or changed by exchanging mesons with other nucleons. If such effect is not negligible, the type of the beta-decay of a free neutron should be different from that of a neutron in a nucleus. The relative magnitude between the Gamow-Teller and Fermi coupling constants, which has been obtained by the analyses of *ft*-values of a neutron and the other light nuclei, probably includes both of the two types of the mesic corrections, one of which exists even in the beta-decay of a free nucleon and the other comes from the many-body effect.

Though it is very difficult to estimate the latter effect quantitatively as well as the former, one can obtain an estimate by noticing that the effects have an intimate relation to the quenching effect for the nuclear magnetic moment as shown in this paper. The corrections to the nuclear magnetic moment due to the interaction between the nucleon current and the electromagnetic field have recently been calculated,<sup>4</sup> using the dispersion relation and assuming that only one pion is exchanged. Analogously it is possible to calculate the many-body corrections to the beta-interaction, also assuming that the one-

pion exchange makes a main contribution. This assumption implies that it is sufficient to replace the  $z$ -component of an isotopic spin operator,  $\tau_3$ , which appears in the calculation of the nuclear magnetic moment, by the other component,  $\tau_+$  or  $\tau_-$ , in order to obtain our results.

The numerical results in section 3, in which we adopt the Fermi gas model as nuclear model, will be discussed in the last section.

## § 2. The many-body mesic corrections and the exchange magnetic moment

Feature of the mesic correction is that the lepton parts of beta-interaction do not suffer any effect, so that the relativistic transformation properties and selection rules of the nucleon parts are invariant. (It can never occur, for example, that an additional pseudoscalar interaction appears from the scalar interaction.) Furthermore, so far as the non-relativistic approximation for the nuclear matrix element can be allowed as the first approximation, there exists no difference between the mesic corrections to the vector and scalar interactions (or the axialvector and the tensor interaction) in the allowed beta-decay.

In this paper we assume that the main part of the many-body effect comes from the one-pion exchange process as shown in Fig. 1a. From the general theorem<sup>4)</sup> it can be proved that the effect of the exclusion principle to the meson cloud in the case of no-spin exchange can be legitimately calculated as the exchange terms of the higher-order process such as Fig. 1a. For simplicity we assume the nucleus to be a closed core and a freely moving nucleon around it. Although the first assumption is sufficiently good, our results may be changed considerably if we take into account the possible strong two-body correlation in the realistic wave function of the nucleus. Therefore the numerical results of next section should not be taken too seriously, but should be understood qualitatively.

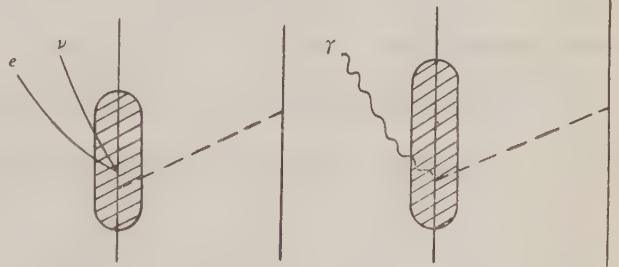


Fig. 1a

Fig. 1b

The nonrelativistic nuclear matrix elements for the allowed beta-decay are conventionally written as  $\int \tau_{\pm}$  (Fermi type) and  $\int \tau_{\pm} \sigma$  (Gamow-Teller type). If we replace  $\tau_{\pm}$  in the vertex operator of Fig. 1a by  $\tau_3$ , we obtain the corresponding process as shown in Fig. 1b, which represents the exchange nuclear magnetic moment due to the electromagnetic interaction with the nucleon current. Recently Fig. 1b has been studied by Hara and one of the authors,<sup>5)</sup> who related the electromagnetic interaction to the pion-nucleon scattering data using the dispersion relation and assuming the validity of the static approximation. As will be noticed at once, only

the Gamow-Teller nuclear matrix element  $\int \tau_{\pm} \sigma$  is corrected by the mesic effect in this paper, because the Fermi matrix element,  $\int \tau_{\pm}$ , cannot lead the nucleon to the isobaric state of total spin  $3/2$  and isospin  $3/2$ , which is prevalent in the cross section of the  $(\pi, \pi)$  scattering or  $(\gamma, \pi)$  production.

### § 3. Detail of the calculation

By the same method as in reference 3 the correction terms corresponding to Fig. 1a to the transition operators of the Gamow-Teller type in the allowed beta-decay are written as :

$$\begin{aligned}\mathfrak{M} &= \sum_{k=1}^6 \sum_{(ij)} \mathfrak{M}_{ij}^{(k)} \\ \mathfrak{M}_{ij}^{(1)} &= (\tau^i + \tau^j)_{\pm} \left\{ \frac{3\mathbf{r}(\mathbf{r} \cdot \boldsymbol{\sigma}^i + \boldsymbol{\sigma}^j)}{r^2} - (\boldsymbol{\sigma}^i + \boldsymbol{\sigma}^j) \right\} f_1(x_{ij}) \mathfrak{M}_l \\ \mathfrak{M}_{ij}^{(2)} &= (\tau^i + \tau^j)_{\pm} (\boldsymbol{\sigma}^i + \boldsymbol{\sigma}^j) f_2(x_{ij}) \mathfrak{M}_l \\ \mathfrak{M}_{ij}^{(3)} &= (\tau^i - \tau^j)_{\pm} \left\{ \frac{3\mathbf{r}(\mathbf{r} \cdot \boldsymbol{\sigma}^i - \boldsymbol{\sigma}^j)}{r^2} - (\boldsymbol{\sigma}^i - \boldsymbol{\sigma}^j) \right\} f_3(x_{ij}) \mathfrak{M}_l \\ \mathfrak{M}_{ij}^{(4)} &= (\tau^i - \tau^j)_{\pm} (\boldsymbol{\sigma}^i - \boldsymbol{\sigma}^j) f_4(x_{ij}) \mathfrak{M}_l \\ \mathfrak{M}_{ij}^{(5)} &= (\tau^i \times \tau^j)_{\pm} \left\{ \frac{3\mathbf{r}(\mathbf{r} \cdot \boldsymbol{\sigma}^i \times \boldsymbol{\sigma}^j)}{r^2} - (\boldsymbol{\sigma}^i \times \boldsymbol{\sigma}^j) \right\} f_5(x_{ij}) \mathfrak{M}_l\end{aligned}$$

and

$$\mathfrak{M}_{ij}^{(6)} = (\tau^i \times \tau^j)_{\pm} (\boldsymbol{\sigma}^i \times \boldsymbol{\sigma}^j) f_6(x_{ij}) \mathfrak{M}_l, \quad (1)$$

where the lepton part

$$\mathfrak{M}_L = i\bar{\psi}_e \gamma_5 \gamma_{\mu} \psi_{\nu} \quad (\text{axialvector}) \quad \text{or} \quad \bar{\psi}_e \gamma_{[\mu\nu]} \psi_{\nu} \quad (\text{tensor}),$$

and

$$\begin{aligned}f_1(x) &= f_3(x) = 2f_5(x) = - (2/15\pi) gA(0) \left( \frac{3}{x^3} + \frac{3}{x^2} + \frac{1}{x} \right) e^{-x} \\ f_2(x) &= f_4(x) = f_6(x) = - (2/15\pi) gA(0) \frac{e^{-x}}{x}\end{aligned} \quad (2)$$

( $x = \mu r$ ;  $\mu$ : the pion rest mass).

We put  $b=c=\mu=1$  throughout this paper. The value of  $A$  for zero-point energy,  $A(0)$ , has relation to the total cross section,  $\sigma_{33}$ , of the pion scattering in the pure  $(3/2, 3/2)$  state as follows :

$$A(0) = (5/18\pi) \int_0^{\infty} (dp/\omega_p^2) \sigma_{33} = 0.33 \quad (3)$$

where the small contributions from  $\sigma_{11}$  and  $\sigma_{13}$  are neglected.

For simplicity we first assume for the nucleus to consist of a closed core plus an odd nucleon, only the latter of which takes the responsibility of the beta-transition. Then the transition matrix element due to the many-body effect is given by

$$(\psi_f, \mathcal{M}\psi_i) = \sum_{i,k} \int (\bar{\psi}_1'(1) \bar{\psi}_i(i) - \bar{\psi}_i(1) \bar{\psi}_1'(i)) \times \mathcal{M}_{i1}^{(k)} \psi_1(1) \psi_i(i) d\mathbf{x}_1 d\mathbf{x}_i, \quad (4)$$

where the transition occurs between the states,  $\psi_1$  and  $\psi_1'$ , and  $\psi_i$  represents a state in the closed core.

It is quite easy to estimate the direct terms of the right-hand of eq. (4), which turn out to be an order of magnitude larger than the exchange terms. The sum of the ordinary Gamow-Teller nuclear matrix elements and the effective one-body corrections arising from the direct terms are expressed numerically as :

$$\mathcal{M}_{\text{ord}} + \mathcal{M}_{\text{dir}} = g(1 - 0.084) \int \tau_{\pm} \sigma. \quad (5)$$

Eq. (5) shows that in the Fermi gas model the Gamow-Teller matrix element is quenched by about 8% owing to the one-pion exchange effect.

For the exchange terms of eq. (4) the numerical calculations can be carried out in a tedious but straightforward way. Then we can express the result as

$$\mathcal{M}_{\text{exch}} = -0.0084g \int \tau_{\pm} (3\mathbf{k}(\sigma \cdot \mathbf{k})/p_F^2 - \sigma), \quad (6)$$

where we assumed, for convenience of the calculation, that the magnitude of the momentum,  $\mathbf{k}$ , of the outside nucleon is just equal to the Fermi momentum,  $p_F$ . If we take off this simplifying assumption, the numerical factor 0.0084 will be slightly modified. Smallness of  $\mathcal{M}_{\text{exch}}$  means that the previous treatment for the mesic effect is almost unchanged, although we take into account the Pauli principle. Apparently  $\mathcal{M}_{\text{exch}}$  depends on the property of the odd nucleon state. To be acquainted with the dependence on the angular momentum of the last nucleon, we calculate the Gamow-Teller nuclear matrix element explicitly in the case where the last nucleon has the total angular momentum  $j$  and the orbital angular momentum  $l$ .

$$(j' || \mathcal{M} || j) = (j' || \mathcal{M}_{\text{ord}} + \mathcal{M}_{\text{dir}} + \mathcal{M}_{\text{exch}} || j), \quad (7)$$

$$(j' || \mathcal{M}_{\text{dir}} || j) = -0.084 (j' || \mathcal{M}_{\text{ord}} || j), \quad (8)$$

$$(j || \mathcal{M}_{\text{exch}} || j) = 0.0084 (j || \mathcal{M}_{\text{ord}} || j) \begin{cases} (2j-1)/2(j+1) & \text{for } j=l+\frac{1}{2} \\ (2j+3)/2j & \text{for } j=l-\frac{1}{2} \end{cases} \quad (9)$$

and

$$(j+1 || \mathcal{M}_{\text{exch}} || j) = -(j || \mathcal{M}_{\text{exch}} || j+1) = -0.0042 (j+1 || \mathcal{M}_{\text{ord}} || j) \quad \text{for } l=j+\frac{1}{2} \quad (10)$$

where  $(j' || \mathcal{M} || j)$ 's are the reduced matrix elements. Table 1 shows the numerical results



for the total corrections to the ordinary nuclear matrix element in the shell model.  $(j' || \mathcal{M}_{\text{ord}} || j)$ .

Table I. Values of  $\mathcal{A}$ 's, which is defined by  $(j' || \mathcal{M} || j) = (1 - \mathcal{A})(j' || \mathcal{M}_{\text{ord}} || j)$

$f \backslash i$	$s_{1/2}$	$p_{3/2}$	$p_{1/2}$	$d_{5/2}$	$d_{3/2}$
$s_{1/2}$	8.4 (%)	—	—	—	—
$p_{3/2}$	—	8.1	8.8	—	—
$p_{1/2}$	—	8.8	5.0	—	—
$d_{5/2}$	—	—	—	7.9	8.8
$d_{3/2}$	—	—	—	8.8	6.7

It is also interesting to show to how much extent the beta-transition can occur in the  $l$ -forbidden case by virtue of the exchange terms. Similar calculations to the case of eq. (9) or (10) lead to

$$(j+1 || \mathcal{M}_{\text{exch}} || j) = -0.0084 \times 3 \sqrt{\frac{(2j+1)(2j+3)}{(j+1)}} \tag{11}$$

where

$$j = l_i - \frac{1}{2}$$
$$j+1 = l_f - \frac{1}{2},$$

if we put the initial and final orbital angular momentum to be  $l_i$  and  $l_f$ . It is clear that the above matrix element is about  $10^4$  times small compared with the ordinary superallowed transition. There are a number of allowed transitions having anomalously long lives, of which the most remarkable example is  $\text{C}^{14} \rightarrow \text{N}^{14} (0^+ \rightarrow 1^-)$  with  $\log ft = 9.0$ , in spite of the fact that  $\log ft = 2.9$  for  $\text{He}^6 \rightarrow \text{Li}^6 (0^+ \rightarrow 1^+)$ . Though some of these transitions may be caused mainly by such mesic effect, it is quite difficult to decide whether the transition arises from the mesic effect or the mixing of the impurity states.

§ 4. Results and discussions

In this paper we have studied the many-body mesic corrections to the allowed beta-transition. The main results are the following two points:

i) The numerical results in the one-pion exchange and the Fermi gas model show that the effective Gamow-Teller coupling constant in heavy nuclei should be about 8% smaller than in the free nucleon, whereas Fermi's one is not changed. It is already known that the mesic corrections in a free nucleon lead to the apparent increase of the Gamow-Teller coupling constant relative to Fermi's one. In a nucleus the above tendency should be somewhat mitigated by the many-body effect. Namely  $g_{GT}$  should be on the average a slightly decreasing function of mass number. The future precise measurement with the  $ft$ -values of neutron and the other light nuclei will give an insight into this problem.

ii) The exchange terms, which make the allowed-type beta-transition possible also in the  $l$ -forbidden cases, turn out to be small as shown in (6) and (11). Probably the discrimination between the mesic effect and the configuration mixing is impossible. However, another aspect of the two-body mesic effect is that the two-particle transition can occur in addition to the one-particle transition, if we stand on the view-point of the independent particle model. The order of magnitude of the former type transition probability is maybe the same as  $6.4 \times 10^{-3}$  times the superallowed transition. It seems to be possible for some of the allowed unfavoured and  $l$ -forbidden transition to be explained by the two-particle transition.

Of course the numerical values in this paper should not be understood quantitatively, since the following additional effects must be also taken into account:

i) The expectation value of the two-body operator is quite sensitive to the existence of the two-body correlation in the nuclear wave function. For example, the deuteron model probably enlarges our effect.

ii) Validity of the static approximation is quite doubtful if two nucleons approach too near. In reference 3 the detailed calculations were carried out, the higher Fourier components being cut off, but the order of magnitude of the numerical results is invariant.

iii) As the next approximation the two-pion effect should be taken into account. The similar problem with the exchange magnetic moment has been treated<sup>2)</sup> by the fourth-order perturbation method, which suggests that the two-pion exchange effect is not so large compared with the one-pion exchange so far as we take the cut-off momentum not so high.

iv) Considerable contribution to the nuclear magnetic moment comes from the Villars' type process as shown in Fig. 2a. If the following process,

$$\pi^+ \rightarrow \pi^0 + e^+ + \nu,$$

exists even in a slight amount,<sup>6)</sup> although the  $\pi - \pi$  decay is not observed by virtue of the smallness of the  $\pi^\pm - \pi^0$  mass difference, another many-body mesic effect of Villars' type as shown in Fig. 2b will appear additionally.

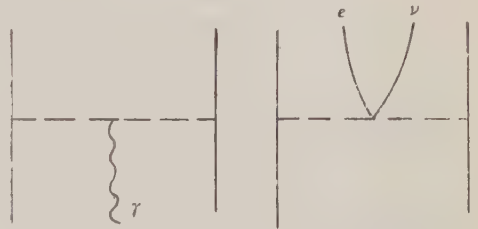


Fig. 2a

Fig. 2b

## § 5. Acknowledgement

The essential idea of this paper was born of the discussion with Doctors T. Katoh, Y. Koh, M. Nozawa, Y. Yoshizawa and T. Nakano, during a visit by one of the authors (Fujita) to Osaka University last summer. He is very grateful to their hospitality and stimulating discussions.

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## Note added in proof

After having written this short note, we received Nuclear Physics Vol. 6, in which we found a paper written by Bell and Blin-Stile with respect to the mesonic effect. Their paper seems to be based on the idea similar to ours, but do not involve the theoretical treatment. Though the meson-theoretical work is referred to in it, it seems to be somewhat different treatment from ours.

## Spontaneous Fission Half-lives

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A dynamical theory of nuclear distortion leading fission, is presented. This theory is based on the unified model (the collective model). But, since the spontaneous fission is, so to speak, an irreversible process, the present theory is treated reflecting this speciality.

Starting from this dynamical theory, a semi-empirical formula of spontaneous fission half-lives of even-even nuclei is derived. This formula can well reproduce the general tendency of spontaneous fission half-lives of even-even nuclei. The essential features which affect half-lives are discussed.

Finally, a reasoning for the asymmetry character of fission and the orientation in calculating the difference of half-lives between even-even and even-odd nuclei are given.

### § 1. Introduction

The theory of nuclear fission has developed in a close relationship with the theory of nuclear structure and nuclear reaction. Bohr and Wheeler<sup>1)</sup> attempted to disclose the mechanism of nuclear fission by making use of the liquid drop model. In those days, the liquid drop model of nuclear structure was regarded to be significant as a compatible model to the compound nucleus theory of nuclear reaction. Then it was considered that nuclear fission should be treated with the same model.

In recent years, the nuclear shell model<sup>2)</sup> succeeded in explaining the ground state spin and magnetic moment of nuclei. It was pointed out by Mayer<sup>3)</sup> that the abundance of fission yield is closely related with magic numbers.

More recently, it was shown that the unified model<sup>3),4)</sup> taking into account both single particle and collective aspects of nucleus can explain the nuclear quadrupole moments and the properties of excited states of nuclei with great success. In this connection some authors<sup>5),6),7),8)</sup> discussed the relation between the shell structure under the deformed nuclear surface and the fission barrier, and the stability of odd parity deformation.

The success of the unified model in explaining nuclear structure, together with the recognition of well organized single particle modes in nuclear reaction<sup>9)</sup> revealed the inadequacy of the liquid drop model. And it is becoming more and more apparent that the liquid drop model is not efficient enough to explain nuclear fission. Inadequacies of applying this model to fission phenomena have been recognized especially in the following two aspects. This model cannot explain

- (1) asymmetric fission, and
- (2) systematics of spontaneous fission half-lives.

Frankel and Metropolis<sup>10)</sup> showed that the liquid drop model with uniform density



is more favourable to the symmetric fission than the asymmetric one. It can also be shown that the asymmetric fission is probable under certain density distribution even in the liquid drop model.<sup>11)</sup> But, on this standpoint, it can be said that any liquid drop model<sup>12)</sup> contradicts with the requirement of the parity conservation. Let us take an example. The ground state of even-even nuclei has even parity. If we wish to explain the asymmetric fission of this type of nuclei with the liquid drop model, we are forced to assume that odd parity states become mixing to even parity states in the course of fission. This is unreasonable.

Huizenga<sup>13)</sup> pointed out two systematics in spontaneous fission half-lives:

(2.1) half-lives of even-odd nuclei is longer than that of corresponding even-even nuclei, and

(2.2) spontaneous fission half-lives of the even-even isotopes correlate with mass number regularly (see Fig. 2 of the present paper). These regularities are two-fold:

(2.2a) for an isotope series of a given element, spontaneous fission half-lives of even-even nuclei go through a maximum with increasing mass number, and

(2.2b) roughly speaking, spontaneous fission half-lives  $\tau$  of the even-even nuclei correlate with mass number  $A$  by a simple relation,

$$\tau = c \cdot \exp(-kA^1),$$

where  $k$  is a constant independent of  $A$  and  $Z$ , and  $c$  is related to  $A$  and  $Z$  simply.

The liquid drop model fails to explain these systematics. That is, this model predicts that the fission half-life becomes longer with increasing  $A$  for fixed  $Z$ . This result contradicts with the experiments.

Thus it is clear that the liquid drop model is too simple to treat nuclear fission. On the other hand, it is apparent that the shell model is also too simple to treat nuclear fission, because the collective aspects of nuclear matter are essential in this phenomenon.

On the contrary, one can understand at least qualitatively (1)<sup>7)</sup> and (2.1),<sup>13)</sup> on the basis of the unified model. And one can expect to explain (2.2) on the same basis. Before entering into discussion about (2.2), however, it would be necessary for us to notice that the fission phenomenon includes two sorts of problems. One is a sort of problems which can be treated well by the kinematical considerations. Angular distributions of photo-fission<sup>14)</sup> and induced fission belong to this sort. The other is a sort of problems which require the dynamical considerations. Explanation of features in spontaneous fission belongs to this sort, because this problem is essentially a dynamical one, being concerned in an irreversible phenomenon. In this view-point, several attempts so far presented,<sup>15-17)</sup> which were based on, so to speak, the "static" considerations, seem to be unsuitable in applying to the spontaneous fission half-lives. This will be criticized in more detail in the last section of the present paper. Rather, we think that the problem should be treated with the time dependent consideration, as was done by Hill and Wheeler<sup>1)</sup> (their Appendix Fig. 34). It was these circumstances which has motivated the present work. In the present paper, consequently, our attention is mainly concentrated to (2) and especially (2.2).

In § 2, a dynamical theory of nuclear distortion leading to fission is presented. In this section, first we shall discuss the reason why the consideration presented by Hill and Wheeler, is applicable to the half-lives of spontaneous fission. The derivation of the equation of Hill-Wheeler is made in Appendix of the present paper. Secondly, the consequences of this equation in applying to spontaneous fission half-lives are presented.

Developing the discussion made in § 2, a semi-empirical formula of spontaneous fission half-lives of even-even nucleus is derived in § 3. This formula can reproduce the general tendency of spontaneous fission half-lives of even-even nuclei well, as will be seen in Fig. 2 and Table 1. The essential features which affect the half-lives are discussed in § 4. Finally a reasoning on the asymmetry character of fission and the orientation in calculating the difference of half-lives between even-even and even-odd nuclei are given.

## § 2. Physical image of the mechanism for spontaneous fission

In this section, we will discuss what plays the important role in determining the spontaneous fission half-lives. This argument leads us to use the equation of Hill-Wheeler. For a while, we will mainly argue the problem of even-odd nuclei for simplicity. But the same argument is also applicable to even-even nuclei. This will be discussed in more detail. Then, we will derive the expression giving the half-lives of spontaneous fission for even-even nuclei. (eq. (11)).

In the even-odd nuclei, it is confirmed that the picture of the single particle moving in a deformed potential well is nicely fitted to the experimental result of nuclear spectroscopy.<sup>17)</sup> Schematically, the energy surface for the single particle in a deformed potential is represented in Fig. 1.

Considering time dependently, the transition probability at a crossing point between two single particle energy surfaces,  $a$  and  $b$ , is decided by three factors<sup>4)</sup>; the transition matrix element,  $H_{ab}$ , the time rate of deformation,  $\dot{\alpha}$ , and the difference in slope of the two unperturbed potential energy surfaces,  $d(E_a - E_b)/d\alpha$ .

The jump probability,  $J_{ab}$ , is very small when  $H_{ab}$  is very weak, or when  $\dot{\alpha}$  is very large, or when  $d(E_a - E_b)/d\alpha$  is very large, or when two or three of these are appreciable simultaneously. At some crossing points,  $H_{ab}$  vanishes owing to the selection rule.  $\dot{\alpha}$  is very large near the equilibrium deformation and is very small near the turning point

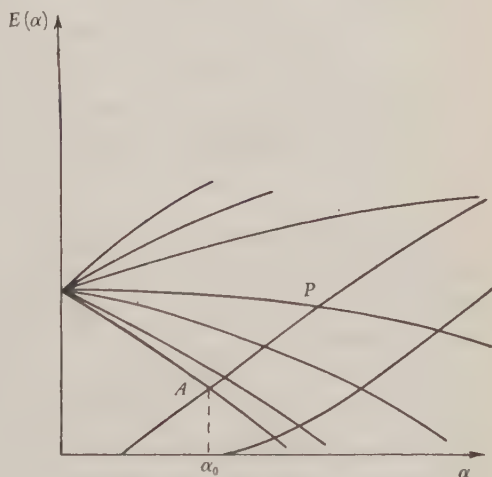


Fig. 1. Schematic representation of the effect of deformation leading to fission upon single particle energy levels.  $\alpha_0$  indicates the equilibrium deformation, where the total energy is minimum with respect to  $\alpha$ .

in semi-classical sense. About  $d(E_a - E_b)/d\alpha$  we will discuss later in this section in more detail.

The jump probability,  $J_{ab}$ , becomes very large when the above three factors contribute constructively. And, at this point  $J_{ab}$  is much larger than  $J_{aa}$ . We name the crossing point satisfying the above condition the typical point "P".

Near the point  $P$ , the jump probability,  $J_{ab}$ , will be effectively contributed by the coupling of two levels,  $E_a$  and  $E_b$ , though  $J_{ab}$  may generally be influenced from many other levels.

Following Hill-Wheeler's expression, the jump probability,  $J_{ab}$ , is given as follows,<sup>4)</sup>

$$J_{ab} = 1 - J_{aa} = 1 - e^{-\pi G^2}, \quad (1)$$

where

$$G^2 = \frac{|H_{ab}|^2}{\hbar \dot{\alpha} d(E_a - E_b)/2d\alpha}. \quad (2)$$

It is expected that the deformation corresponding to  $P$  will not be larger than the one corresponding to the fission threshold in a semi-classical sense. However, after passing the typical crossing point  $P$  once, the single particle energy  $E(\alpha)$  is fed into the surface oscillation energy. Then  $\dot{\alpha}$  becomes larger and larger, the jump probability becomes smaller and smaller at further crossing points and the deformation proceeds on. Consequently, the half-lives of spontaneous fissions seem to be decided by the jump probability,  $J_{ab}$ , at the typical point  $P$ . Hence the half-lives are given by

$$\tau = J_{ab}^{-1}. \quad (3)$$

In the above argument, we assume the single extra-particle for simplicity. This situation, that the single particle moves in a deformed potential, is justified by the recent experiment.<sup>17)</sup> This guarantees the validity of the adiabatic approximation adopted in the above arguments.

In the case of two extra particles, there are no direct experimental proofs of the validity of the adiabatic approximation as yet. But under the following situation, we could expect the applicability of the same approximation. According to the shell model, the nuclei in this region are characterized by the large  $j$ , the spin of the single particle state. Following Mayer's argument,<sup>21)</sup> the pairing energy for the extra particles is large for large  $j$ . This will cause the situation that the direct coupling between extra particles over-whelms the particle-surface coupling and consequently the extra particles are coupled to the surface in the same manner as a single particle ( $BM^{3,2})$  II, c)).

Hence the argument of this section will be applied without a serious modification also to the even-even nuclei. That is, the above discussion does not set the limits to the even-oddness of the nuclei.

In what follows, we limit our attention to the even-even nuclei, since in this sort of nuclei slow and regular variations of the various experimental quantities are expected whereas in the even-odd nuclei abrupt changes of these quantities are expected from nucleus to nucleus.<sup>17)</sup>

In the region considered,  $|H_{ab}|^2$  also may not depend so much on mass number, etc. Since in the very neighborhood of equilibrium  $\dot{\alpha}$  is very large,  $J_{ab}$  becomes so small that the slippage does not occur effectively. In other words,  $\dot{\alpha}$  is very small and it is taken to be constant with mass number etc., at the crossing point slipping effectively, irrespective of the order of magnitude of the zero point energy of nuclear surface oscillation. Accordingly, the probability of slippage is affected only by the difference between the slopes of energy surfaces  $E_a(\alpha)$  and  $E_b(\alpha)$ . This can be estimated roughly as follows. Expanding  $E(\alpha)$  to the second order of  $\alpha$ , we put

$$E(\alpha) = E^{(0)} + E^{(1)} \cdot \alpha + E^{(2)} \cdot \alpha^2. \quad (4)$$

As for the mass number dependence of  $E^{(2)}$ , we adopt the one given by Bohr and Wheeler.<sup>1)</sup>

Then,

$$E^{(2)} \propto A^{2/3} \cdot (1-x),$$

$$x = (Z^2/A) / (Z^2/A)_{\text{crit.}}, \quad (5)$$

$$(Z^2/A)_{\text{crit.}} = 44.65 \quad (\text{using the value given by Fermi}^{19}).$$

And then, we may assume the following relation.

$$E_a(\alpha) - E_b(\alpha) = \rho(A) \cdot (K^{(0)} + K^{(1)} \cdot \alpha + K^{(2)} \cdot \alpha^2 A^{2/3} \cdot (1-x)), \quad (6)$$

where  $K^{(0)}$ ,  $K^{(1)}$  are constants, i. e., independent of the mass number, and  $\rho(A)$  is the function of  $A$ . Assuming the nucleus as the potential well with finite depth,  $\rho(A)$  can be found to be a decreasing function of  $A$ , since the difference in the slopes of the energy levels of unperturbed single particle states become smaller with increasing mass number. And  $K^{(1)}$  is not always smaller than  $K^{(2)}$  since  $K^{(1)}$  and  $K^{(2)}$  correspond to the difference between  $E_a^{(1)}$  and  $E_b^{(1)}$ , and  $E_a^{(2)}$  and  $E_b^{(2)}$ , respectively, though  $E^{(1)}$  itself is generally much smaller than  $E^{(2)}$  in the neighbourhood of the closed shell. The region under consideration is far from the closed shell, so  $E^{(1)}$  does not vanish.

In order to estimate  $\alpha$  in eq. (6) from experimental quantity, we use the relation  $E_{z,p.} \propto 1/J$ , where  $E_{z,p.}$  is the energy of zero point oscillation. And  $J$  is the moment of inertia which is related to  $\alpha$  as  $J \propto M_\alpha \cdot \alpha^2$ , where,  $M_\alpha$  is the effective mass for nuclear capillary oscillation.

$E_{z,p.}$  can be estimated from the equation

$$E_{z,p.} = (1/2) \cdot (\text{the experimental value of the energy of the first excited state}).$$

Hence we get

$$\alpha = K \sqrt{1/M_\alpha \cdot E_{z,p.}} = K' A^{-5/6} \cdot E_{z,p.}^{-1/2}, \quad (7)$$

where  $K$  and  $K'$  are constants with the change of mass number. Expanding eq. (2) to the power of  $\alpha$ , we get,

$$G \cong \frac{|H_{ab}|}{\sqrt{\hbar \dot{\alpha}}} \cdot \frac{1}{\sqrt{\rho(A)}} (K^{(1)} - K^{(2)} \alpha \cdot A^{2/3} (1-x)). \quad (8)$$

Using eqs. (1) and (3),



$$\begin{aligned}\log_{10} \tau &\simeq \log_{10} (1 - e^{-\pi G^2})^{-1} \\ &\simeq e^{-\pi G^2}.\end{aligned}\quad (9)$$

Because  $J_{ab} \gg J_{aa}$ ,  $G$  is very large. Therefore the above expression is appropriate. Further in expanding the last expression, we take some  $G_i$ , expand it to the first order of  $G - G_0$  and get

$$\log_{10} \tau = A - B \cdot G, \quad (10)$$

where  $A$  and  $B$  are some constants.  $G_i$  is the average value of  $G$  at the typical crossing point over the nuclei under consideration. Combining eqs. (8) and (10), we get finally  $\log_{10} \tau$  as follows.

$$\log_{10} \tau = \mathfrak{A} - \mathfrak{B}' \cdot [\rho(A)]^{-1/2} + \mathfrak{C} [\rho(A)]^{-1/2} \cdot A^{-1/6} (1 - x) E_{7,0}^{-1/2}, \quad (11)$$

where,  $\mathfrak{A}$ ,  $\mathfrak{B}'$  and  $\mathfrak{C}$  are positive constants and determined approximately as adjustable parameters with experimental values of known three pairs of half-life and the energy of the first excited state. Now there are 13 nuclei for which both the half-life of the spontaneous fission and the energy of the first excited state are measured and so these 13 nuclei are studied by means of three adjustable parameters.

### § 3. Derivation semi-empirical formula

W. J. Swiatecki<sup>17)</sup> gave the semi-empirical formula giving the fission half-lives in connection with mass number, but his formula does not seem to stand on a sufficient theoretical basis. In this section we will derive the semi-empirical formula following the arguments developed in Section 2.

For  $\rho(A)$  of eq. (11), it is tempting to assume a decreasing function of  $A$ , having a form of  $\rho(A) = \gamma^2 A^2$  ( $\gamma$ ; constant). This would not be so unreasonable in the region considered here. Then, eq. (11) becomes

$$\log_{10} \tau = \mathfrak{A} - \mathfrak{B} A + \mathfrak{C} A^{5/6} \cdot (1 - x) \cdot E_{7,0}^{-1/2}, \quad (12)$$

where

$$\mathfrak{B} = \mathfrak{B}' \cdot \gamma.$$

The above form well indicates the typical features of the experimental results of spontaneous fission half-lives. In the case that the second term is large compared with the third term, really it will be shown in later numerical calculation that the ratio of the third term to the second is about 15-100, the main features of the half-lives are determined by the first and the second term. And the third term appears as the correction to the formers. Actually, it is well known that the relation

$$\log_{10} \tau = \mathfrak{A} - \mathfrak{B} \cdot A \quad (13)$$

represents the gross behavior of the mass number dependence of spontaneous fission half-lives. This is the relation which was stated in (2.2b) of the introduction of this paper. The experimental values of energies of first excited states, being adopted here as the measure of the zero point energy, have some regularity, as is well known.<sup>18)</sup> And we

can find that this regularity has some correlation with experimental data for half-lives of even-even nuclei, that is, in U and Pu the energy of the first excited state goes through a minimum with increasing mass number for given  $Z$ . The nuclei showing the above minimum correspond to those having the maximum point in the curves of spontaneous fission for even-even nuclei (see Fig. 2).

Adopting  $\mathcal{A}=102$ ,  $\mathcal{B}=0.441$  and  $\mathcal{C}=5.76$  for  $\mathcal{A}$ ,  $\mathcal{B}$  and  $\mathcal{C}$ , respectively ( $E_{z,n}$  is measured in unit of kev.), the result is given in Fig. 2 and Table 1. In this result, the calculated values of Th and U are rather small in comparison with the experimental results. However, we may conclude that this result represents well the general tendency of experimental data.

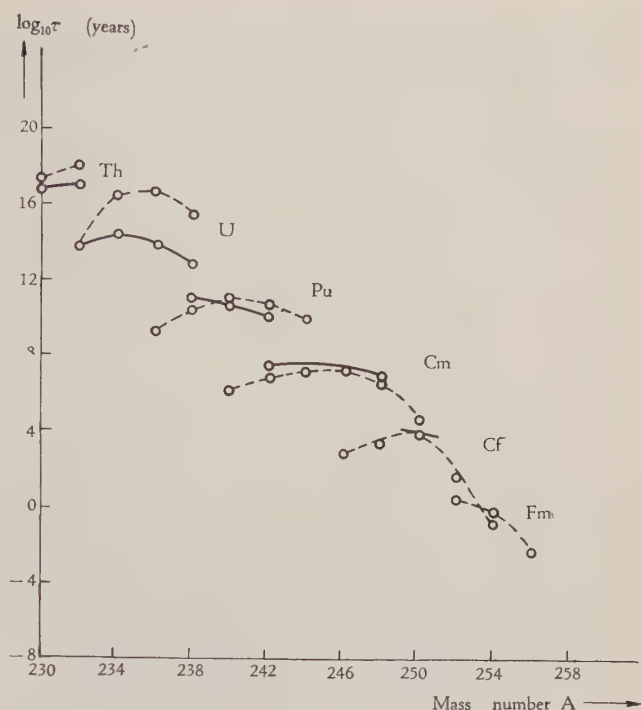


Fig. 2. Mass number dependence of half-lives in spontaneous fission. The dotted curve represents experimental results. The solid curve represents the calculated value using eq. (13).

#### § 4. Discussion and concluding remarks

In spite of much simplification, our model seems to be able to reproduce the complex experimental results consistently. If we want to study this problem in more detail, we have to estimate  $|H_{nn}|^2$ , and the contribution from lower levels. At present, we have not enough knowledge to calculate these factors in detail. These knowledges should include more reasonable estimate of effective mass of capillary oscillation than that predicted by the present liquid drop model. Further, it is necessary to have more information about the structure of matrix elements and so on. The factor  $\rho(A)$  in the preceding section must be estimated by using the deformed potential with finite depth, though we assumed here an inverse square proportionality with mass number for it.

However, taking these into account makes not only the calculation much complicated but also the insight ambiguous. As a result it is considered that the conclusions in § 2 and § 3 indicate the essential quality of this phenomena. Namely the half-lives of the spontaneous fission are decided by the probability of slippage in the crossing point of

Table 1. Half-lives of spontaneous fission

Z	A	E (kev)	log <sub>10</sub> τ (years)	
			Calculated values	Experimental values
90	230	52.4	16.3	17.2
	232	49.0	16.0	18.2
92	232	47.0	14.1	13.9
	234	43.5	14.4	16.3
	236	45.2	13.9	16.3
	238	50.0	12.9	15.9
94	238	44.1	11.0	10.7
	240	42.9	11.1	11.1
	242	44.6	10.5	10.9
	244	42.3	7.9	6.86
96	246	42.9	7.4	7.48
	248	43.4	7.1	6.62
98	250	42.0	4.1	4.18

E; the energy of the first excited state.

For Th, I. Perlman and J. O. Rasmussen, *Alpha Radioactivity*, Encyclopedia of Physics vol. XIII, Nuclear reactions III (1957), Springer-Verlag (Berlin).

For U, P. C. Allen, Phys. Rev. **105** (1957), 1796.

For other nuclei, E. K. Hyde and G. T. Seaborg, *The transuranium Elements*, Encyclopedia of Physics, Vol. XLII, Nuclear reactions III (1957), Springer-Verlag (Berlin).

τ; half-lives of spontaneous fission.

For Pu<sup>242</sup>, Mech et. al., Phys. Rev. **103** (1956), 340.

For Cm<sup>248</sup>, Butler et. al., Phys. Rev. **103** (1956), 965.

For others, those values are used which are collected by A. Ghiorso and cited by W. J. Swiatecki (Phys. Rev. **100** (1955), 937). These values are in accord with E. K. Hyde and G. T. Seaborg (l. c. p. 306) except Th<sup>232</sup>.

the energy levels, at which the three factors  $H_{\alpha}$ ,  $\alpha$  and  $d(E_{\alpha} - E_{\beta})/d\alpha$  appearing in (2) work constructively, and especially influenced by the difference in the slopes of the energy levels with reference to the deformation.

Our treatment is different from those attempts<sup>(1-3)</sup> to calculate the potential barrier for fission by the static considerations and to estimate the penetration probability for it. In these trials, the probability of slippage in crossing points is always assumed to be unity so far as the whole levels are closely occupied by particles in order from the bottom of the potential well. In these models, it is assumed that the unoccupied levels never come down below the uppermost occupied level in any stage of the deformation. But such an assumption cannot be justified and within such a restriction we may not expect to obtain good results in calculating spontaneous fission half lives. Naturally, at the initial stage of the transition probability  $J_{\alpha\beta}$  from a level  $\alpha$  to another level  $\beta$  varies with energies of zero point oscillation, which show some regular variation upon mass number as remarked in the previous section. Consequently,  $J_{\alpha\beta}$  should not always be equal to unity. In our treatment, on the contrary, this situation is taken into account properly.

Finally, we will discuss the asymmetry character in nuclear fission and the difference of half-lives between even-even nuclei and even-odd nuclei. First we shall discuss the asymmetry character. The level distance between the closed shell and the upper levels of the unperturbed (i. e. not so deformed) nucleus is very large. Consequently, the crossing points of closed shell and the upper levels appear in far distorted form compared to the initial distortion. This circumstance gives rise to the following two situations. One is that the number of crossing points near the initial stage of the distortion will be much reduced. Hence, at the decisive stage of spontaneous fission which is expected to lie not so far from the initial distortion, the closed shell nature is not so disturbed. The other feature is that near the closed shell the difference of slopes of concerning levels is large in the neighborhood of the crossing points. This makes the transition probability  $J_{ab}$  small at these points in this region compared with that at crossing points in other region. As a result, the levels below closed shell tend to conserve the single particle nature in deformed potential. That is, even in the middle stage of the process these levels may conserve the closed shell nature fairly well. If the collective oscillation of odd parity is raised in this stage, as a result of excitation of several particles coupled together with odd parity, the asymmetric character in fission will appear. Hereby asymmetric fission and closed shell nature will be strongly correlated. In such a stage, however, we must take into account many concerned levels and the adiabatic approximation may not be justified. In this case, the quantitative treatment of fission asymmetry have much complexity.

As Newton<sup>17)</sup> pointed, it may be considered that the difference in half-lives between the even-even nuclei and even-odd nuclei reflects the following circumstances. In heavy nuclei, generally the total angular momentum of single particle is so large that the pairing energy is also large. And two particles are easy to couple together. Then, in even-even nuclei each particle is able to jump over from the initial level to the next level with either the same parity or the different parity from the initial one. On the other hand, in even-odd nuclei, when one particle outside the core carries the parity of the nucleus, the requirement of parity conservation forbids this particular particle to move to another level with different parity. Of course, this argument is more complicated when several particles are coupled together and the states resulting the coupling of these particles include both even and odd parity. However, as is mentioned in § 2, it is confirmed experimentally<sup>17)</sup> that the single particle moving in a deformed potential is the suitable model in even-odd nuclei, this argument is to be taken as reasonable. According to the above considerations we can understand that the lives of even-odd nuclei are longer than those of even-even nuclei. A corresponding character of even-odd nuclei is found in alpha decay.<sup>21)</sup> Quantitative discussion of the problem requires, however, the more detailed knowledge of matrix element  $H_{ab}$ .

In conclusion, authors express their sincere gratitude to Professor M. Kobayasi for his kind advices throughout this work.



### Appendix

In this appendix, we will derive Hill-Wheeler's equation (Fig. 34 in their Appendix),<sup>4)</sup> starting from the total Hamiltonian of the system.

Now we consider the point,  $Q$  where two single particle energy surfaces ( $a$  and  $b$  in Fig. 3) are crossing each other. Here the contributions from other levels ( $c$  in Fig. 3) are assumed to be neglected.

Now, the Hamiltonian of the total system is given as follows,

$$H = H_p(x) + H_{\text{int}}(x, \alpha) + H(\alpha) + H'(x, \alpha). \quad (\text{A} \cdot 1)$$

Here,

$$H_p(x) + H_{\text{int}}(x, \alpha) + H(\alpha) \quad (\text{A} \cdot 2)$$

is the Hamiltonian proposed by A. Bohr,<sup>3)</sup> where  $H_p$  is the Hamiltonian of an extra particle,  $H(\alpha)$  is that of the surface oscillation, including up to the square of  $\alpha$ , and  $H_{\text{int}}(x, \alpha)$  is that of the interaction between the single particle and the surface oscillation.  $H'(x, \alpha)$  represents the residual interactions including all the remaining part, which was neglected by A. Bohr.  $H'(x, \alpha)$  includes the third and higher powers of  $\alpha$ , besides the interactions between nucleons probably, which could be expressed as the body oscillation, and so forth. Then, this part may include other characteristic coordinates, for instance, the one describing the body oscillation mode, etc., but generally it can be written by using  $x$  and  $\alpha$  because of the conservation of degree of freedom.

Inclusion of  $H'(x, \alpha)$  in (A.1) is necessary for a system which undergoes spontaneous fission. The Schrödinger equation of the total system is

$$i\hbar \frac{\partial \Psi}{\partial t} = H\Psi. \quad (\text{A} \cdot 3)$$

We will solve the above equation with time dependent perturbation method, initial condition being

$$\Psi = \psi_a(x, \alpha) \cdot f_a(\alpha). \quad (\text{A} \cdot 4)$$

This initial condition is suitable under nearly adiabatic conditions. This is the so-called perturbed stationary state wave function,<sup>22)</sup> which is already perturbed by  $H(\alpha)$ , but  $\alpha$  is treated as though  $\alpha$  were fixed.  $\psi_a$  is the eigen-solution of the equation

$$(H_p + H_{\text{int}}(x, \alpha)) \cdot \psi_a(x, \alpha) = W_a(\alpha) \cdot \psi_a(x, \alpha). \quad (\text{A} \cdot 5)$$

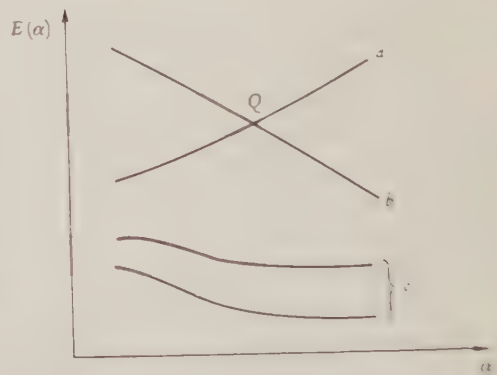


Fig. 3. Schematic representation of single particle energy surfaces.

The functions  $\psi'_m(x, \alpha)$  form an ortho-normal set with respect to  $x$  for all values of the parameter  $\alpha$ .  $f_\alpha(\alpha)$  is the eigen-solution of

$$H(\alpha) \cdot f_\alpha(\alpha) = \varepsilon_\alpha \cdot f_\alpha(\alpha). \quad (\text{A} \cdot 6)$$

In order to solve eq. (A.3), the following interaction representation is adopted.

$$\begin{aligned} \Psi &= e^{-i(H_p + H_{\text{int}})t/\hbar} \Psi^I(x, \alpha, t) \\ H^{II} &= e^{i(H_p + H_{\text{int}})t/\hbar} \cdot H' \cdot e^{-i(H_p + H_{\text{int}})t/\hbar}. \end{aligned} \quad (\text{A} \cdot 7)$$

Then eq. (A.3) is transformed as follows,

$$i\hbar(\partial/\partial t)\Psi^I = H^{II}\Psi^I, \quad (\text{A} \cdot 8)$$

and the initial condition becomes as

$$\Psi^I(t = -\infty) = \Psi. \quad (\text{A} \cdot 9)$$

Near the crossing point  $Q$ , we can put  $\Psi^I$  as

$$\Psi^I = \psi'_a \cdot g_a(\alpha, t) + \psi'_b g_b(\alpha, t), \quad (\text{A} \cdot 10)$$

where

$$g_m(\alpha, t) = \int \psi_m^*(x, \alpha) \Psi^I(x, \alpha, t) dx. \quad (\text{A} \cdot 11)$$

Introducing eq. (9) into eq. (7), we get the simultaneous equations,

$$\begin{aligned} i\hbar \dot{g}_a &= H_{aa}^I g_a + H_{ab}^I g_b \\ i\hbar \dot{g}_b &= H_{ba}^I g_a + H_{bb}^I g_b, \end{aligned} \quad (\text{A} \cdot 12)$$

where

$$H_{mn}^I = \int \psi_m^* (H(\alpha) + H'(x, \alpha)) \psi_n \cdot dx \cdot e^{i(W_m(\alpha) - W_n(\alpha))t/\hbar}. \quad (\text{A} \cdot 13)$$

As for the diagonal part of eq. (A.12), we put

$$H_{nn}^I = \int \psi_n^* (H(\alpha) + H'(x, \alpha)) \psi_n dx = E_n(\alpha) \quad (\text{A} \cdot 14)$$

and as for the non-diagonal part, we put

$$H_{mi}^I = \int \psi_m^* H'(x, \alpha) \psi_n dx e^{i(W_m(\alpha) - W_n(\alpha))t/\hbar} = H_{mi}. \quad (\text{A} \cdot 15)$$

Then eq. (A.12) with eqs. (A.14) and (A.15) is the equation of Hill and Wheeler.

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## Effects of Defects on Lattice Vibrations

———*Thermodynamical Properties\**———

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An attempt is given for calculating the thermodynamical quantities with respect to the vibrational frequencies of the imperfect lattices. Using the method of contour integral the expressions for the difference of vibrational free energies between imperfect and perfect lattices are derived. These are given in two forms available at high and at low temperatures.

As the examples of the practical calculations using these formulae, the  $n$ -dimensional simple lattices with defects are treated. Especially the simple cubic lattice as well as the linear chain are investigated in detail. Although in this example a simplified model of the actual defect is considered, it may be enough to discuss the general features of the thermodynamical properties of the imperfect lattices.

### § 1. Introduction

The calculation of the free energy of the crystal with defects has been done for the Schottky or Frenkel defects, although it was only a phenomenological one. Recently the investigation of the influence of lattice imperfections such as impurities, holes, and dislocations on the physical properties of crystals seems to have been one of the subjects of great current interest in solid state physics. The thermodynamical properties of the crystal with defects also have been investigated by several authors.<sup>1-3)</sup>

First, Kirkwood and Stripp<sup>1)</sup> calculated the influence of the Schottky defects on the vibrational part of the crystal partition function by means of perturbation theory. Their evaluation of the partition function seems, however, to be fairly approximate because only the first term of the expansion in terms of  $T^{-1}$ ,  $T$  being the temperature, is retained. In the problem of the evaluation of the effect of defects on the vibrational partition function there may appear two difficulties: the first, the troublesome problem of the evaluation of the influence of defects on the frequency spectra of the lattice vibration and the second, the difficulty to carry out the summation over all the normal modes.

The problem of the first kind has been treated by several authors<sup>2, 4-10)</sup>. Most of

\* This work was started while one of the authors (T. Tanaka) was at the University of Maryland and was completed after he returned to the Kyusyu University. Quite recently we learned that the same problem had been taken up by the Maryland group (J. Mahanty, A. Maradudin and G. Weiss) after T. Tanaka came back to Japan and that a similar result was obtained (Prog. Theor. Phys., this issue). Since the two groups had developed the formulation rather independently until they obtained the final results, it was hoped that the two results be published in the same place.



them have limited their considerations only to the one dimensional lattice, while the general theory developed by Montroll and Potts<sup>2</sup> is applicable to the case of three dimensional lattice and, actually, a general expression for the additive functions of the frequencies of normal mode vibrations of the crystal lattices has been given without perturbation. The second difficulty, however, seems to remain still unsolved. Montroll, Potts, and others<sup>2,6,7</sup> calculated the zero-point energy by means of replacing the sum by the contour integral on the frequency-plane; nevertheless, the difficulty of integration seems to have limited the range of possible calculation remarkably.

On the contrary, in the case of the vibrational part of the crystalline free energy the method of contour integral seems to be more convenient than the usual perturbation because, fortunately, the special form of the function of the free energy (logarithmic form) makes possible the integration of the contour integral on the frequency-plane. (See § 2). Although our method is, of course, not applicable to the investigation as to the effect of defects on the other parts of the crystalline free energy, it is rather useful so far as the vibrational free energy is concerned.

In § 2 the expression for the imperfect lattice is derived by the method of contour integral. Since this is of the integrated form, we can find the final result at once if the characteristic function—that is, the secular determinant for determining the normal frequencies of the imperfect lattice divided by that of the perfect one—is given in any way as the function of frequencies. This expression is, however, of the form that is convenient at moderately high temperatures. Therefore the expression available at low temperatures is derived next. This is of the form such that it consists of the change of the zero-point energy from the perfect lattice and the temperature dependent correction terms against this. Then, the methods for calculating the characteristic functions are reviewed in § 3. The method of Montroll and Potts<sup>2</sup> is employed in this paper for evaluating the characteristic functions.

As the examples of the practical calculations using these formulae, the  $n$ -dimensional simple lattices with nearest neighbor interactions are investigated. The discussion is confined mainly to the case of impurities including isotopes. In addition only the harmonic terms are, for simplicity, retained in the potential, that is, the effects of thermal expansion and any relaxation of the structure about the defects are ignored. In § 4 the change of the vibrational free energy due to one defect will be considered. The calculations are done for the cases of the simple cubic lattice, monatomic and diatomic linear chains. The interactions between defects and that between a defect and the boundaries are investigated in § 5. Then, the influence of many disordered defects on the vibrational free energy is treated in § 6 and is shown that the method of Montroll and Potts<sup>2</sup> for determining the characteristic functions is also useful in some degree to the case of many disordered defects.

## § 2. General formulae for the free energy of the imperfect lattice

First, we consider the quantity:

$$G = \sum_j g(\omega_j) \quad (2.1)$$

where  $g(\omega)$ 's are arbitrary functions of frequency  $\omega$  and the summation is extended over all the normal frequencies  $\omega_j$ 's of the lattice vibrations. In the present paper we shall use, for convenience, as the frequency and temperature the reduced (dimensionless) quantities

$$\omega \equiv \omega/\omega_L \text{ and } T \equiv 2\pi(\kappa T/\hbar\omega_L), \quad (2.2)$$

where  $\omega_L$  is the maximum frequency of the perfect lattice and  $\kappa$  is the Boltzmann constant. Since the normal frequencies  $\omega_j$ 's are the solutions of the secular equation,

$$D(\omega) = (\text{const}) \cdot \prod_j (\omega^2 - \omega_j^2) = 0, \quad (2.3)$$

we can write (2.1) in the form

$$G = \frac{1}{2\pi i} \oint_{C'} g(\omega) d[\log D(\omega)] \quad (2.4)$$

as we get from (2.3)

$$d[\log D(\omega)] = \sum_j \left\{ \frac{1}{\omega - \omega_j} + \frac{1}{\omega + \omega_j} \right\} d\omega. \quad (2.5)$$

In (2.4), the contour  $C'$  is such that it includes all the positive zero points of  $D(\omega)$  in it and not any singular point of  $D(\omega)$  and  $g(\omega)$ . Therefore, when the secular equation of normal frequencies of the perfect lattice is given in the form

$$D_0(\omega) = (\text{const}) \cdot \prod_j (\omega^2 - \omega_j^{02}) = 0, \quad (2.6)$$

we obtain as the difference quantity  $\Delta G$  of  $G$  between the imperfect and perfect lattices:

$$\Delta G = G - G_0 = \frac{1}{2\pi i} \oint_C g(\omega) d \left[ \log \frac{D(\omega)}{D_0(\omega)} \right]. \quad (2.7)$$

In (2.7) the contour  $C$  is similar to the  $C$ 's in (2.4). Hence, if the function  $g(\omega)$  is regular on the positive half  $\omega$ -plane we can take as  $C$  the following; that is, it consists of the positive semi-circle of radius  $R$  and the vertical diameter along the imaginary axis on the  $\omega$ -plane. Since  $d \log(D(\omega)/D_0(\omega))$  is given as the difference of two quantities; one is given by (2.5) and the other is the corresponding quantity of the perfect lattice, we can prove that the contribution of the semi-circle to the integral (2.6) becomes zero as  $R$  tends to infinity if the function  $g(\omega)$  behaves  $\sim |\omega|^X$  ( $2 > X \geq 0$ ) as  $|\omega| \rightarrow \infty$ ; namely,

$$\begin{aligned} \frac{1}{\omega - \omega_j} + \frac{1}{\omega + \omega_j} &= \frac{2\omega}{\omega^2 - \omega_j^2}, \\ \frac{2\omega}{\omega^2 - \omega_j^2} - \frac{2\omega}{\omega^2 - \omega_j^{02}} &= 2\omega \frac{\omega_j^2 - \omega_j^{02}}{(\omega^2 - \omega_j^2)(\omega^2 - \omega_j^{02})} \sim O\left(\frac{1}{\omega^3}\right) \end{aligned}$$

$$\oint_R g(\omega) \cdot O\left(\frac{1}{\omega^3}\right) d\omega \sim \frac{|\omega|^X}{\omega^2} \rightarrow 0 \quad (\text{if } 2 > X \geq 0) \quad \text{as } |\omega| \rightarrow \infty.$$

Then the contribution of the integral in (2.7) comes only from the integral along the imaginary axis if the function  $g(\omega)$  has such a property. Furthermore, noticing that  $D(\omega)$  and  $D_0(\omega)$  are the function of  $\omega^2$ ,  $d \log(D(\omega)/D_0(\omega))$  is an odd function on the imaginary axis. We can, then, conclude that only the odd part of  $g(\omega)$  contributes to the integral in (2.7) on the imaginary axis. That is,

$$\Delta G = -\frac{1}{\pi i} \int_0^\infty [g(i\omega')]_{\text{odd}} \frac{d}{d\omega'} \left[ \log \frac{D(i\omega')}{D_0(i\omega')} \right] d\omega'. \quad (2.8)$$

If we put  $g(\omega) = \frac{1}{2} \hbar \omega$ , we obtain as the difference of the zero-point energy  $\Delta E$

$$\Delta E_0 = -\frac{\hbar \omega_L}{2\pi} \int_0^\infty \omega' \frac{d}{d\omega'} \left[ \log \frac{D(i\omega')}{D_0(i\omega')} \right] d\omega'. \quad (2.9)$$

Eq. (2.9) is the same as that obtained by Montroll-Potts.<sup>2</sup> (Hereafter we refer to it as (I).)

Next, we consider the Helmholtz free energy of the system of harmonic oscillators. Since it is given by

$$F = \frac{1}{\beta} \sum_j \log [2 \sinh \left( \frac{\pi}{T} \omega_j \right)]; \quad \beta = \frac{2\pi}{\hbar \omega_L} \frac{1}{T}, \quad (2.10)$$

we may put

$$g(\omega) = \frac{1}{\beta} \log \left[ 2 \sinh \left( \frac{\pi}{T} \omega \right) \right].$$

Then, since

$$g(i\omega') = \frac{1}{\beta} \left\{ \log \left[ 2 \sinh \left( \frac{\pi}{T} \omega' \right) \right] \pm i\pi \left( k + \frac{1}{2} \right) \right\}; \quad \omega' \gtrless 0$$

$$\text{for } k\pi \leq \frac{\pi}{T} |\omega'| < (k+1)\pi,$$

we obtain

$$[g(i\omega')]_{\text{odd}} = \pm i\pi \left( k + \frac{1}{2} \right) / \beta; \quad \omega' \gtrless 0$$

$$\text{for } k\pi \leq \frac{\pi}{T} |\omega'| < (k+1)\pi.$$

Inserting this into (2.8), we get

$$\begin{aligned} \Delta F &= F - F_0 \\ &= -\frac{1}{2\beta} \int_0^\infty \frac{d}{d\omega'} \left[ \log \frac{D(i\omega')}{D_0(i\omega')} \right] d\omega' - \frac{1}{\beta} \sum_{k=0}^\infty k \int_{k\pi}^{(k+1)\pi} \frac{d}{d\omega'} \left[ \log \frac{D(i\omega')}{D_0(i\omega')} \right] d\omega'. \end{aligned} \quad (2.11)$$

Eq. (2.11) is integrated at once and we obtain the final result :

$$\Delta F = \frac{1}{2\beta} [A(0) - A(\infty)] + \frac{1}{\beta} \sum_{k=0}^{\infty} k [A(kT) - A((k+1)T)], \quad (2.12)$$

where

$$A(\omega') = \log \frac{D(i\omega')}{D_0(i\omega')}. \quad (2.12a)$$

Eq. (2.12) is of the form available at high temperature, because the second term becomes zero in the limit of high temperature and so can be regarded as the correction to the first term at high temperature. This can be easily seen by expanding  $D(\omega)$  in terms of  $D_0(\omega)$ . From (2.3) and (2.6)

$$\frac{D(\omega)}{D_0(\omega)} = (\text{const}) \left[ 1 - \sum_j (\omega_j^2 - \omega_j^{02}) \frac{D'_j(\omega^2 - \omega_j^2)}{D_j(\omega^2 - \omega_j^{02})} + \dots \right]. \quad (2.12b)$$

Since we may put  $\omega = ikT$  in order to insert  $D(\omega)/D_0(\omega)$  into the second term of (2.12),  $|\omega|$  becomes large at high temperature and the second term of the above expansion is of the order of  $1/|\omega|^2$ , the third term of the order of  $1/|\omega|^4$ , etc., which are of the order of  $1/T^2$ ,  $1/T^4$ , etc., respectively. Then the second term becomes zero as  $T$  tends to infinity.

Although (2.12) can be used at all temperatures, it is very inconvenient below ordinary temperatures; because, at low temperatures, the zero point energy is dominant. Then it is desirable to rewrite (2.12) in the form in which the zero-point energy appears as the first term plus correction terms. Fortunately, this is done very easily using the Euler-Maclaurin summation formula :

$$\begin{aligned} \sum_{k=0}^{\infty} g(k) &= \int_0^{\infty} g(k) dk + \frac{1}{2} [g(0) + g(\infty)] \\ &+ \sum_{l=1}^{\infty} (-1)^l \frac{B_l}{(2l)!} [g^{(2l-1)}(0) - g^{(2l-1)}(\infty)], \end{aligned} \quad (2.13)$$

where  $B_l$  is the Bernoulli number of the  $l$ -th order. If we put

$$g(k) = k[A(kT) - A((k+1)T)]$$

in (2.13) and perform the partial integral,\* we obtain

$$\Delta F = \Delta E_0 + \frac{\hbar \omega_L}{2\pi} \sum_{l=1}^{\infty} T^{2l} \alpha_l(T) \quad (2.14)$$

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\* In the derivation of the expression (2.14) we regard the quantity  $A(kT)$  as that which is normalized so that the constant factor of (2.12b) is 1. The expression (2.14), however, can be used for the quantity  $A(kT)$  which is unnormalized. The authors are indebted to Mr. I. Mannari of Kyoto University who pointed out an error in a previous derivation of the second term of (2.14).



with

$$\alpha_l(T) = (-1)^l \frac{(2l-1)}{(2l)!} B_l \frac{1}{T} [A^{(2l-2)}(0) - A^{(2l-2)}(T)] \\ - \frac{1}{(2l+1)!} A^{(2l-1)}(0) - \frac{T}{(2l+2)!} A^{(2l)}(0), \quad (2.14a)$$

where  $\mathcal{J}E_0$  is given by (2.9). Eq. (2.14) is the low temperature expansion for  $\mathcal{J}F$  and is the second result we have wished to derive.

If the series  $\sum A(kT)$  converges, the second term of (2.14) becomes the expansion in terms of  $T$ . It is easily seen by the rearrangement of the series  $\sum g(k)$ : namely,

$$\sum_{k=0}^{\infty} g(k) = \sum_{k=0}^{\infty} k [A(kT) - A((k+1)T)] \\ = \sum_{k=1}^{\infty} A(kT).$$

Then, inserting this into (2.13) we obtain

$$\mathcal{J}F = \mathcal{J}E_0 + \frac{\hbar\omega_L}{2\pi} \sum_{l=1}^{\infty} T^{2l} \alpha'_l \quad (2.15)$$

with

$$\alpha'_l = (-1)^l \frac{B_l}{(2l)!} A^{(2l-1)}(0). \quad (2.15a)$$

It is worth noting that (2.12) is of the integrated form. In other words, we have only to calculate the characteristic function  $D(\omega)$   $D_i(\omega)$  at any rate, and the difference of free energy  $\mathcal{J}F$  is obtained at once by (2.12). The second term of (2.12), however, converges very slowly in general. Hence, it is more convenient to expand it in terms of the power series of  $1/T^2$ . In most cases it is performed easily.

Finally, we consider the quantity  $\mathcal{J}C_r$ , that is, the difference of the heat capacities between the perfect and imperfect lattices. It is obtained at once from  $\mathcal{J}F$  by the following expression.

$$\mathcal{J}C_r = -2\pi \frac{\kappa T}{\hbar\omega_L} \left( \frac{\partial^2}{\partial T^2} \mathcal{J}F \right). \quad (2.16)$$

From (2.12) and (2.14)  $\mathcal{J}C_r$  is given in the form of series. When  $\mathcal{J}F$  is given in the form of series in terms of  $1/T^2$  or  $T$ ,  $\mathcal{J}C_r$  is, of course, obtained in the same form.

### § 3. The characteristic functions

In this section we shall review briefly the method of calculating the characteristic functions developed in (I) for the convenience of the later calculation.

In  $n$ -dimensional lattice, the time-independent part of the equations of motion of the atom at the lattice point  $\mathbf{s} = (s_1, s_2, \dots, s_n)$  are given by

$$\omega^2 \omega_L^2 M_{\mathbf{s}} u_{\mathbf{s}}^k + \sum_{\mathbf{s}', l} \Gamma_{\mathbf{s}\mathbf{s}'}^{k,l} u_{\mathbf{s}'}^l = 0 \quad \left( \begin{array}{l} k, l=1, 2, \dots, n \\ \mathbf{s}, \mathbf{s}'=1, 2, \dots, N \end{array} \right), \quad (3.1)$$

where  $\mathbf{u}_{\mathbf{s}} = (u_{\mathbf{s}}^1, u_{\mathbf{s}}^2, \dots, u_{\mathbf{s}}^n)$  is the time-independent part of the displacement vector of the atom  $\mathbf{s}$  from its equilibrium position and  $\Gamma(\mathbf{s}, \mathbf{s}'; k, l)$  is the component of the "force constant tensor".

If we use as the characteristic functions,  $D(\omega)$ ,  $D_0(\omega)$ , the determinants eliminating all the  $\mathbf{u}_{\mathbf{s}}$  from the equations of motion of the imperfect and perfect lattices, respectively, these are given in the following forms:

$$\begin{aligned} D_0(\omega) &= \det[a(\mathbf{s}, k; \mathbf{s}', l)], \\ D(\omega) &= \det[a(\mathbf{s}, k; \mathbf{s}', l) + \Delta(\mathbf{s}, k; \mathbf{s}', l)], \end{aligned} \quad (3.2)$$

$$\left( \begin{array}{l} k, l=1, 2, \dots, n \\ \mathbf{s}, \mathbf{s}'=1, 2, \dots, N \end{array} \right).$$

The determinants  $D(\omega)$  and  $D_0(\omega)$  are of the  $nN^n$ -th order when the total number of the lattice points is  $N^n$ . If the number of the defects is small compared with that of the normal atoms, we can expand  $D(\omega)$  in terms of  $D_0(\omega)$  and obtain

$$\frac{D(\omega)}{D_0(\omega)} = \det[\delta_{\mathbf{s}, \mathbf{k}; \mathbf{s}', l} + \sum_{(\mathbf{s}'', q)} \Delta(\mathbf{s}, k; \mathbf{s}'', q) a^{-1}(\mathbf{s}', l; \mathbf{s}'', q)]. \quad (3.3)$$

The right-hand side of (3.3) is the determinant of  $p$ -th order if the maximum number of non zero  $\Delta$  in the row of  $D(\omega)$  is  $p$  and the summation of  $(\mathbf{s}'', q)$  is taken over all the values of  $\mathbf{s}''$  and  $q$  for which  $\Delta(\mathbf{s}, k; \mathbf{s}'', q) \neq 0$ . The  $a^{-1}(\mathbf{s}, k; \mathbf{s}'', q)$  is the  $(\mathbf{s}, k; \mathbf{s}'', q)$  element of the inverse matrix of  $D_0(\omega)$  and is given by the following expression:

$$a^{-1}(\mathbf{s}, k; \mathbf{s}', l) = \sum_{\phi_j=1}^N [\lambda^l(\phi_j)]^{-1} u_{\mathbf{s}'}^l * (\phi_j) u_{\mathbf{s}}^k(\phi_j), \quad (3.4)$$

where  $\lambda^l(\phi_j)$  and  $u_{\mathbf{s}'}^l(\phi_j)$  are the eigenvalue and corresponding eigen function of (3.1).

We now consider, for simplicity, only the nearest neighbour interaction. Then eq. (3.1) becomes

$$\begin{aligned} \omega^2 \omega_L^2 M_{s_1 s_2 \dots s_n} \mathbf{u}_{s_1 s_2 \dots s_n} \\ + \sum_{k=1}^n \gamma_k^l [\mathbf{u}_{s_1 s_2 \dots s_k - 1 \dots s_n} + \mathbf{u}_{s_1 s_2 \dots s_k + 1 \dots s_n} - 2\mathbf{u}_{s_1 s_2 \dots s_k \dots s_n}] = 0, \end{aligned} \quad (3.5)$$

where  $\gamma_k^l$  is the force constant associated with the displacement of the atom parallel to the  $k$ -th coordinate axis. Fortunately, each equation of (3.5) has the same form for different values of  $l$  and in the equation of motion of the  $l$ -th component of  $\mathbf{u}$ , there appear only the  $l$ -th's and not other's. Then, if we rearrange the rows and columns of  $D(\omega)$  and  $D_0(\omega)$  appropriately, they become the product of  $n$  determinants of  $N^n$ -th order; that is

$$D_0(\omega) = \prod_{j=1}^n D_0^j(\omega) = \prod_{j=1}^n \det_j [a_j(s, s')],$$

$$D(\omega) = \prod_{j=1}^n D^j(\omega) = \prod_{j=1}^n \det_j [a_j(s, s') + \mathcal{A}_j(s, s')], \quad (3.6)$$

where

$$a_j(s, s') = \omega^2 \omega_L^2 M_s \partial_{s, s'} + \sum_{l=1}^m \gamma_l^j (\partial_{s_l'} s_l - 1 + \partial_{s_l'} s_l + 1 - 2\partial_{s_l'} s_l) \prod_{k=1}^n \partial_{s_k'} s_k, \\ (k \neq l)$$

Therefore, the difference of free energies of imperfect and perfect lattices is given in the form

$$\mathcal{J}F = \sum_{j=1}^n \mathcal{J}F^j$$

and the result of § 2 holds for each  $\mathcal{J}F^j$ . Since we have only to discuss any one of  $\mathcal{J}F^j$ s we hereafter omit the affix  $j$  for brevity.

#### § 4. Free energy of the lattice with one defect

4/1)  $n$ -dimensional monatomic lattice.

We consider as the defect the following three models: (see eq. (3.1))

A) Impurity

$$M_s \rightarrow M_{s'}, \quad \Gamma_{s, s'}^{k, l} \rightarrow \Gamma_{s, s'}^{l, k}.$$

B) Isotopic impurity (Isotope)

$$M_s \rightarrow M_{s'}.$$

C) Hole

$$\gamma \rightarrow \gamma' \quad (\text{only one place}) \quad \text{when } n=1.$$

Clearly, B) is a special case of A). Since it may be physically improper to regard the hole as an impurity which is characterized as the limiting case of  $M \rightarrow 0$  and  $\gamma \rightarrow \gamma'$ , we temporarily take the above model only in the linear chain.

Now we consider the case in which only one impurity exists in the perfect monatomic lattice of  $n$ -dimension. Then  $(2n+1)$  equations become different from (3.5). These are

$$[\omega^2 \omega_L^2 M_{s_1 \cdots s_k \pm 1 \cdots s_n} - (2\gamma_1 + \cdots + \gamma_k + \gamma_k' + \cdots + 2\gamma_n)] \cdot u_{s_1 \cdots s_k \pm 1 \cdots s_n} \\ + \sum_{l=1}^n \gamma_l \cdot [u_{s_1 \cdots s_k \pm 1 \cdots s_l - 1 \cdots s_n} + u_{s_1 \cdots s_k \pm 1 \cdots s_l + 1 \cdots s_n}] \\ (l \neq k) \\ + [\gamma_k \cdot u_{s_1 \cdots s_k \pm 2 \cdots s_n} + \gamma_k' \cdot u_{s_1 \cdots s_k \cdots s_n}] = 0, \quad (k=1, 2, \cdots, n)$$

$$\begin{aligned} & [\omega^2 \omega_L^2 \mathbf{M}'_{s_1 \dots s_k \dots s_n} - \sum_{k=1}^n 2\gamma'_k] \cdot \mathbf{u}_{s_1 \dots s_k \dots s_n} \\ & + \sum_{k=1}^n \gamma'_k \cdot [\mathbf{u}_{s_1 \dots s_k - 1 \dots s_n} + \mathbf{u}_{s_1 \dots s_k + 1 \dots s_n}] = 0. \end{aligned} \quad (4.1)$$

The characteristic function  $D(\omega)/D_0(\omega)$  is, therefore, the determinant of the  $(2n+1)$ -th order of which the form is (3.3). This is rather complicated to treat and so we consider the following two cases: (i) the simple cubic lattice and (ii) the isotopic impurity in  $n$ -dimensional lattice.

(i) *Simple cubic lattice*

In this case the characteristic function is the determinant of the seventh order given in Appendix. Since the total number of lattice points,  $N^3$ , is very large, the sum in (3.4) can be approximately replaced by the integral. (That is, neglecting the order of  $O(1/N)$ .) Furthermore, if we use the Born-von Kármán condition, eq. (3.4) becomes

$$a^{-1}(\mathbf{s}, \mathbf{s}'; i\omega') = \frac{-1}{(2\pi)^n} \int_0^{2\pi} \dots \int_0^{2\pi} \frac{\prod_{l=1}^n \cos[(s_l - s'_l) \theta_l] d\theta_l}{\omega'^2 \omega_L^2 M + \sum_{l=1}^n 2\gamma_l (1 - \cos \theta_l)}. \quad (4.2)$$

Eq. (4.2) is rewritten by the simple transformation<sup>11)</sup> into the following expression.

$$a^{-1}(\mathbf{s}, \mathbf{s}'; i\omega') = - \int_0^\infty dx \exp[-x (\sum_{l=1}^n 2\gamma_l) (1 + 2\omega'^2)] \prod_{l=1}^n I_{s_l - s'_l}(2\gamma_l x). \quad (4.3)$$

Hereafter, we consider the isotropic case for simplicity; that is,  $\gamma_1 = \gamma_2 = \dots = \gamma_n = \gamma$ . Of course, our result can be extended to the non-isotropic case by means of a slight modification. Then (4.3) becomes

$$a^{-1}(\mathbf{s}, \mathbf{s}'; i\omega') = - \frac{1}{2\gamma} P_n(\mathbf{s} - \mathbf{s}'; \omega'), \quad (4.4)$$

where

$$P_n(\mathbf{s} - \mathbf{s}'; \omega') = \int_0^\infty dx \exp[-nx(1 + 2\omega'^2)] \prod_{l=1}^n I_{s_l - s'_l}(x). \quad (4.5)$$

The characteristic function given in Appendix is calculated easily using these functions. The result is

$$\begin{aligned} \frac{D(i\omega')}{D_0(i\omega')} &= [1 - \delta'(\rho_1 + \rho_3)]^3 [1 - \delta'(\rho_1 + 2\rho_2 - \rho_3)]^2 \\ &\times [1 - \epsilon(2n\omega'^2)P_0 - \delta'(7\rho_1 - 4\rho_2 - \rho_3) \\ &+ \epsilon\delta' \{ (2n\omega'^2 P_0)(\rho_1 - 4\rho_2 - \rho_3) + 6(2n\omega'^2 P_1)\rho_2 \} \\ &+ 6\delta'^2(\rho_1 - \rho_2)(\rho_1 - 4\rho_2 - \rho_3)] \end{aligned} \quad (4.6)$$

where



$$\epsilon = 1 - M'/M, \quad \delta' = 1 - \gamma' / \gamma, \quad (4.7)$$

and

$$\begin{aligned} \rho_1 &= \frac{1}{2} \{P_3(000; \omega') - P_3(001; \omega')\}, \\ \rho_2 &= \frac{1}{2} \{P_3(001; \omega') - P_3(011; \omega')\}, \\ \rho_3 &= \frac{1}{2} \{P_3(001; \omega') - P_3(002; \omega')\}, \\ P_0 &\equiv P_3(000; \omega'), \quad P_1 \equiv P_3(001; \omega'). \end{aligned} \quad (4.8)$$

Furthermore, remembering the following properties of the function  $P(s-s'; \omega')$ :

$$2n\omega'^2 P_n(s-s'; \omega') \rightarrow 0 \quad (\omega' \rightarrow 0) \quad (4.9)$$

$$2n\omega'^2 P_n(s-s'; \omega') \rightarrow \delta_{s,s'} \quad (\omega' \rightarrow \infty)$$

$$P_3(000; 0) = 0.4990$$

$$P_3(001; 0) = 0.1658$$

$$P_3(002; 0) = 0.0778$$

$$P_3(011; 0) = 0.1045,$$

(4.10)\*

we obtain for the free energy difference,  $\Delta F$ , owing to one impurity

$$\Delta F = \frac{1}{2\beta} \log G_0(\epsilon, \delta') + \frac{1}{\beta} \sum_{k=0}^{\infty} k \log \frac{G(kT; \epsilon \delta')}{G((k+1)T; \epsilon \delta')} \quad (4.11)$$

where

$$G_0(\epsilon, \delta') = (1-\epsilon)^{-1} (1-\delta') (1-0.2106\delta')^3 (1-0.1839\delta')^2$$

$$G(kT; \epsilon \delta') = \frac{D(ikT)}{D_0(ikT)}. \quad (4.12)$$

At higher temperatures the first term of (4.11) is dominant, so the vibrational free energy decreases owing to one impurity if it satisfies the following condition:

$$G_0(\epsilon, \delta') < 1,$$

or, from (4.7),

$$\frac{M}{M'} \frac{\gamma'}{\gamma} (0.7894 + 0.2106 \frac{\gamma'}{\gamma})^3 (0.8161 + 0.1839 \frac{\gamma'}{\gamma})^2 < 1. \quad (4.13)$$

Now, consider the expansion of the second term of (4.11) in terms of  $1/T^2$  at sufficiently high temperatures. Since from (2.12) and (2.12a)  $\omega' = kT$  in the second term of (4.11),  $\omega'$  becomes large at higher temperatures. Therefore, the integrand of

\* The values in (4.10) are in accordance with reference 11). Although these values seem to have the errors of some 0.5%, it does not exert any serious influence to our results.

(4.2) can be expanded in terms of  $(2\gamma/M\omega_L^2 \omega'^2) = (2n\omega'^2)^{-1}$  and easily integrated using the orthogonality conditions of the cosine functions; namely,

$$P_n(\mathbf{s}-\mathbf{s}'; \omega') = \left(\frac{1}{2n\omega'^2}\right) \sum_{j=0}^{\infty} \left(\frac{-1}{2n\omega'^2}\right)^j q_j(\mathbf{s}-\mathbf{s}') \quad (4.14)$$

where  $q_j$  is defined by

$$q_j(\mathbf{s}-\mathbf{s}') = \left(\frac{1}{2\pi}\right)^n \int_0^{2\pi} \cdots \int_0^{2\pi} \left[\sum_{l=1}^n (1 - \cos \theta_l)\right]^j \prod_{m=1}^n \cos[s_m - s'_m] d\theta_m. \quad (4.14a)$$

The special values of  $q_j$  for the small values of  $j$  and  $(\mathbf{s}-\mathbf{s}')$  are shown in Table 1. The expansion of the second term of (4.11) using (4.14a) is, however, very tedious

Table 1.

$q_0(0 \cdots 0) = 1,$	$q_1(0 \cdots 0) = n,$	$q_2(0 \cdots 0) = n^2 + \frac{1}{2}n, \cdots$
$q_0(0 \cdots 1) = 0,$	$q_1(0 \cdots 1) = -\frac{1}{2},$	$q_2(0 \cdots 1) = -n, \cdots$
.....		

though straightforward, so we consider the case in which  $\epsilon$  and  $\delta'$  are small. Picking up only the first order of  $\epsilon$  and  $\delta'$ , we get the following expression as the high temperature expansion of  $\Delta F$ .

$$\begin{aligned} \Delta F &= \frac{1}{2\beta} \log G_0(\epsilon, \delta') \\ &+ \frac{1}{\beta} (\epsilon - 2\delta') \left[ \zeta(2) \frac{1}{2T^2} - \left(1 + \frac{1}{2n}\right) \zeta(4) \left(\frac{1}{2T^2}\right)^2 + \left(1 + \frac{3}{2n}\right) \zeta(6) \left(\frac{1}{2T^2}\right)^3 - \cdots \right] \\ &\quad (n=3). \end{aligned} \quad (4.15)$$

Next, we consider the low temperature expansion of  $\Delta F$ . It is obtained at once inserting the characteristic function, (4.6), into eqs. (2.14), (2.14a) and (2.9). The integral of  $\Delta E_0$  however, seems to us too hard to give the exact integration. Therefore, we consider only a special case in which  $\epsilon$  and  $\delta'$  are small. Then the integrand of  $\Delta E_0$ , i.e.  $\log(D(\omega)/D_0(\omega))$ , can be expanded and integrated, because the integration of  $P(\mathbf{s}-\mathbf{s}'; \omega')^m$  with respect to  $\omega'$  is of the Gaussian type; namely,

$$\begin{aligned} &\int_0^{\infty} (2n\omega'^2)^l [P_n(\mathbf{s}-\mathbf{s}'; \omega')]^m d\omega' \\ &= \frac{(2l-1)!!}{2^l} \left(\frac{\pi}{2n}\right)^{1/2} p_{l/2}^{(m)}(\mathbf{s}-\mathbf{s}'), \end{aligned} \quad (4.16)$$

where  $p_{l/2}^{(m)}(\mathbf{s}-\mathbf{s}')$  is a constant number given by the following integral:

$$p_{i/2}^{(m)}(\mathbf{s}-\mathbf{s}') = \int_0^\infty \int (x_1 + \dots + x_m)^{-l/2} e^{-\epsilon \left( \sum_{k=1}^m x_k \right)} \prod_{j=1}^n \prod_{l=1}^m I_{s_j - s_j'}(x_k) dx_k. \quad (4.17)$$

Therefore,  $\Delta E_0$  becomes, for small  $\epsilon$  and  $\delta'$ ,

$$\begin{aligned} \Delta E_0 &\sim \frac{\hbar \omega_L}{2\pi} \int_0^\infty [\epsilon \cdot 2n\omega'^2 P_3(000; \omega') + 6\delta' \{P_3(000; \omega') - P_3(001; \omega')\}] d\omega' \\ &= \frac{\hbar \omega_L}{2\pi} \left[ \epsilon \cdot \frac{1}{4} \left( \frac{\pi}{6} \right)^{1/2} \gamma_{3/2}^{(1)}(000) + 3\delta' \left( \frac{\pi}{6} \right)^{1/2} \{p_{1/2}^{(1)}(000) - p_{1/2}^{(1)}(001)\} \right]. \end{aligned} \quad (4.18)$$

Then, the low temperature expansion of  $\Delta F$  for small  $\epsilon$  and  $\delta'$  becomes as follows:

$$\begin{aligned} \Delta F &= \frac{\hbar \omega_L}{2\pi} \left[ \epsilon \frac{1}{4} \left( \frac{\pi}{6} \right)^{1/2} p_{3/2}^{(1)}(000) + 3\delta' \left( \frac{\pi}{6} \right)^{1/2} \{p_{1/2}^{(1)}(000) - p_{1/2}^{(1)}(001)\} \right] \\ &\quad + \frac{\hbar \omega_L}{2\pi} \sum_{l=1}^\infty T^{2l} \alpha_l' \end{aligned} \quad (4.19)$$

where the second correction terms are calculated easily from (2.15a), namely,

$$\begin{aligned} \alpha_l' &\equiv (-1)^{l-1} \frac{B_l}{(2l)!} \left[ \left( \frac{\partial}{\partial \omega} \right)^{2l-1} \{ \epsilon (2n\omega^2) P_3(000; \omega) \right. \\ &\quad \left. + 6\delta' P_3(000; \omega) - 6\delta' P_3(001; \omega) \} \right]_{(\omega=0)}. \end{aligned} \quad (4.20)$$

## (ii) Isotopic impurity.

The difference of free energy  $\Delta F$  due to one isotope in the simple cubic lattice is obtained at once by putting  $\delta'=0$  in the expansion of (i). However, we can discuss not only the simple cubic lattice but generally the  $n$ -dimension case, because the characteristic function for one isotope is the determinant of the first order.

The characteristic function for one isotope is given by

$$\begin{aligned} D(i\omega') &= 1 - \epsilon 2n\omega'^2 P_n(0 \cdots 0; \omega'). \\ D_0(i\omega') & \end{aligned} \quad (4.21)$$

Inserting (4.21) into (2.12) and with (4.9), we obtain the difference of free energy due to one isotope; namely,

$$\Delta F = \frac{1}{2\beta} \log \frac{1}{1-\epsilon} + \frac{1}{\beta} \sum_{k=0}^\infty k \log \left\{ \frac{1 - \epsilon 2nk^2 T^2 P_n(0 \cdots 0; kT)}{1 - \epsilon 2n(k+1)^2 T^2 P_n(0 \cdots 0; (k+1)T)} \right\}. \quad (4.22)$$

The high temperature expansion of (4.22) can be performed in the same way as was done in (i), while (4.14a) is simplified in the present case. It is necessary to calculate only  $q_j(0 \cdots 0)$ , which is given by

$$q_j(0 \cdots 0) = \sum_{A=0}^{[j/2]} \frac{j!}{(j-2A)!} n^{j-2A} \sum_{(\mu_m)} \prod_m \frac{1}{2^{\mu_m} (\mu_m)!} \quad (4.23)$$

where the summation with respect to  $\mu_m$  is taken over all the sets that satisfy the following condition:

$$\sum_{m=1}^n \mu_m = A.$$

Then, the high temperature expansion of  $\Delta F$  is given by

$$\begin{aligned} \Delta F &= \frac{1}{\beta} \log \left[ (1-\epsilon)^{-1/2} \left\{ 1 - \epsilon \sum_{j=1}^{\infty} q_j (0 \cdots 0) \zeta(2j) \left( \frac{-1}{2nT^2} \right)^j \right\} \right] \\ &= \frac{1}{\beta} \log \left[ (1-\epsilon)^{-1/2} \left\{ 1 + \epsilon \zeta(2) \frac{1}{2T^2} - \epsilon \left( 1 + \frac{1}{2n} \right) \zeta(4) \left( \frac{1}{2T^2} \right)^2 \right. \right. \\ &\quad \left. \left. + \epsilon \left( 1 + \frac{3}{2n} \right) \zeta(6) \left( \frac{1}{2T^2} \right)^3 - \cdots \right\} \right]. \end{aligned} \quad (4.24)$$

In the simple cubic lattice, Eq. (4.24) becomes, of course, the same expression as (4.15) in which we put  $\delta' = 0$  ( $\gamma' = \gamma$ ). In one dimensional case eq. (4.24) becomes

$$\Delta F = \frac{1}{\beta} \log \left[ (1-\epsilon)^{-1/2} \left\{ 1 - \epsilon \sum_{j=1}^{\infty} \binom{2j}{j} \zeta(2j) \left( \frac{-1}{2T^2} \right)^j \right\} \right]. \quad (4.25)$$

It can be easily proved that (4.25) is the same as (4.34) with (4.35b).

The low temperature expansion also is obtained in the same manner as in (i). Inserting (4.21) into (2.14),

$$\Delta F = \frac{\hbar \omega_L}{2\pi} \int_0^{\infty} \log [1 - \epsilon 2n \omega'^2 P_n(0 \cdots 0; \omega')] d\omega' + \frac{\hbar \omega_L}{2\pi} \sum_{l=1}^{\infty} T^{2l} \alpha'_l. \quad (4.26)$$

The integral in (4.26) is performed at once from (4.16) if  $\epsilon$  is small. Then, the result is

$$\Delta F = \frac{\hbar \omega_L}{2\pi} \cdot \epsilon \left( \frac{\pi}{2n} \right)^{1/2} p_{3/2}^{(j)}(00 \cdots 0) + \frac{\hbar \omega_L}{2\pi} \sum_{l=1}^{\infty} T^{2l} \alpha'_l \quad (4.27)$$

where  $\alpha'_l$ 's are calculated from (2.15a) and (4.21) straightforwardly, i.e.,

$$\alpha'_l = (-1)^l \frac{B_l}{(2l)!} \left[ \left( \frac{\partial}{\partial \omega} \right)^{2l-1} \log \left\{ 1 - \epsilon \frac{\omega}{v' \sqrt{1 + \omega^2}} \right\} \right]_{(\omega=0)}. \quad (4.28)$$

Eq. (4.27) is to be compared with eq. (4.19).

From (4.22) and (4.28) the vibrational free energy decreases on account of one isotope if the following condition is satisfied.

$$\epsilon < 0 \text{ or } \frac{M'}{M} > 1. \quad (4.29)$$

Eq. (4.29) corresponds to (4.12). When the normal atom is replaced by the heavier isotope, the normal frequencies of the lattice vibrations may be decreased. So eq. (4.29)



is expected naturally.

#### 4/2) Monatomic linear chain.

Monatomic linear chain is the only case in which the integral of the Green function (4.2) can be performed exactly. Therefore, we shall investigate this particularly.

The Green function of Monatomic linear chain is given by

$$a^{-1}(s, s'; i\omega') = -\frac{1}{2\gamma} P_1(s-s'; \omega') = \frac{1}{\gamma} \frac{z^{1s-s'}}{z-z^{-1}}; \omega' > 0 \quad (4.30)$$

where

$$z = [\sqrt{\omega'^2 + 1} - \omega']^2. \quad (4.31)$$

The characteristic functions of one impurity, one isotope and one hole are calculated easily and the results are (See Appendix II.)

$$\frac{D(i\omega'; \epsilon)}{D_0(i\omega'; \epsilon)} = 1 - \epsilon \frac{\omega'}{\sqrt{1 + \omega'^2}}; \text{ isotope}, \quad (4.32a)$$

$$\frac{D(i\omega'; \delta)}{D_0(i\omega'; \delta)} = 1 - \delta \frac{\omega'}{\sqrt{1 + \omega'^2}}; \text{ hole}, \quad (4.32b)$$

$$\begin{aligned} \frac{D(i\omega'; \epsilon \delta)}{D_0(i\omega'; \epsilon \delta)} &= 1 - (\epsilon + 2\delta) \frac{\omega'}{\sqrt{1 + \omega'^2}} \\ &+ 4\delta\epsilon \frac{\omega'^2}{\sqrt{1 + \omega'^2}} [\sqrt{1 + \omega'^2} - \omega'] \quad (4.32c) \\ &+ 4\delta^2(1 - \epsilon) \frac{\omega'^3}{\sqrt{1 + \omega'^2}} [\sqrt{1 + \omega'^2} - \omega']^2; \text{ impurity}, \end{aligned}$$

where

$$\epsilon = 1 - M'/M, \quad \delta = 1 - \gamma/\gamma' = \frac{-\delta'}{1 - \delta'}. \quad (4.33)$$

In eqs. (4.32b) and (4.32c) we have used the fact that  $\mathcal{J}F$  is invariant even if the constant factor is multiplied to the characteristic function. It seems to be that from (4.32)  $\epsilon$  in the case of isotope,  $\delta$  in the case of hole and  $(\epsilon + 2n\delta)$  in that of impurity are corresponding to each other. This may be due to the properties of our models as defects and, therefore, holds in other cases.

Inserting (4.32) into (2.12) we get the following expression of  $\mathcal{J}F$ .

$$\mathcal{J}F = \frac{1}{2\beta} \log G_0(\epsilon, \delta) + \frac{1}{\beta} \sum_{k=0}^{\infty} k \log \frac{G(\epsilon, \delta; kT)}{G(\epsilon, \delta; (k+1)T)} \quad (4.34)$$

where

$$G_0(\epsilon, \delta) = \begin{cases} (1 - \epsilon)^{-1} & ; \text{ isotope,} \\ (1 - \delta)^{-1} & ; \text{ hole,} \\ (1 - \delta)^{-2} (1 - \epsilon)^{-1} & ; \text{ impurity,} \end{cases} \quad (4.35a)$$

and

$$\begin{aligned}
 & 1 - \epsilon \frac{kT}{\sqrt{1+k^2T^2}} \quad ; \text{ isotope,} \\
 & 1 - \delta \frac{kT}{\sqrt{1+k^2T^2}} \quad ; \text{ hole,} \\
 G(\epsilon, \delta; kT) = & 1 - (\epsilon + 2\delta) \frac{kT}{\sqrt{1+k^2T^2}} \quad (4.35b) \\
 & + 4\delta\epsilon \frac{k^3T^2}{\sqrt{1+k^2T^2}} [\sqrt{1+k^2T^2} - kT] \\
 & + 4\delta^2(1-\epsilon) \frac{k^3T^3}{\sqrt{1+k^2T^2}} [\sqrt{1+k^2T^2} - kT] \\
 & \quad ; \text{ impurity.}
 \end{aligned}$$

From (4.35b) the high temperature expansion of  $\Delta F$  is obtained at once.

$$\Delta F = \frac{1}{2\beta} \log G_0(\epsilon, \delta) + \frac{1}{\beta} \sum_{q=1}^{\infty} f_q T^{-2q}; \quad \text{for } \left| \frac{1}{2G_0-1} \right| > 1. \quad (4.36)$$

The first several values of  $f_q$  are shown in Table 2. The expansion for  $1 > (2G_0-1)^{-1} > 0$  is also obtained in the same manner. For the impurity, eq. (4.36) is correct so far as the first order of  $\epsilon$  and  $\delta$  are concerned.

Above the ordinary temperature the first term of (4.34) becomes dominant. Then the condition that the vibrational free energy decreases owing to one defect is following.

$$G_0(\epsilon, \delta) < 1$$

or

$$\begin{aligned}
 (M'/M) &> 1 && ; \text{ isotope,} \\
 (\gamma/\gamma') &> 1 && ; \text{ hole,} \\
 (M'/M) (\gamma/\gamma')^2 &> 1 && ; \text{ impurity.}
 \end{aligned} \quad (4.37)$$

This corresponding to (4.12) and (4.29).

The low temperature expansion is also obtained easily by inserting (4.32) and (4.34) into (2.15); namely,

$$\Delta F = \Delta E_0 + \sum_{l=1}^{\infty} \alpha_l' T^{2l} \quad (4.38)$$

with

$$\begin{aligned}
 \Delta E_0 = & \left( \frac{\hbar\omega_L}{\pi} \frac{G_0}{\sqrt{2G_0-1}} \tan^{-1} \sqrt{2G_0-1} - \frac{1}{4} \hbar\omega_L; \quad 2G_0-1 \geq 0 \right. \\
 & \left. \frac{\hbar\omega_L}{2\pi} \frac{G_0}{\sqrt{1-2G_0}} \log \frac{1-\sqrt{1-2G_0}}{1+\sqrt{1-2G_0}} - \frac{1}{4} \hbar\omega_L; \quad 1-2G_0 > 1, \right. \\
 & \left. \right) \quad (4.39a)
 \end{aligned}$$

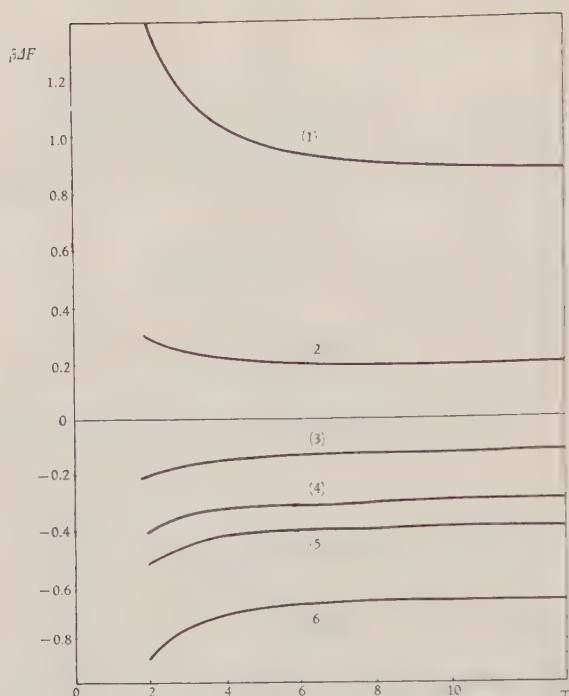


Fig. 1.  $\Delta F$  due to isotope or hole in the monatomic linear chain.

(1)  $\cdots M' = (2/11)M$ , or  $\gamma' = (11/2)\gamma$ . (2)  $\cdots M' = (2/3)M$ , or  $\gamma' = (3/2)\gamma$ . (3)  $\cdots M' = (4/3)M$ , or  $\gamma' = (3/4)\gamma$ . (4)  $\cdots M' = (20/11)M$ , or  $\gamma' = (11/20)\gamma$ . (5)  $\cdots M' = (20/9)M$ , or  $\gamma' = (9/20)\gamma$ . (6)  $\cdots M' = 4M$ , or  $\gamma' = (1/4)\gamma$ .

and the first several values of  $\alpha_i'$  are

$$\alpha_1' = \frac{B_1}{2!} \frac{G_0 - 1}{G_0}, \quad \alpha_2' = \frac{B_2}{4!} \left\{ 3 \frac{G_0 - 1}{G_0} - 2 \left( \frac{G_0 - 1}{G_0} \right)^3 \right\}, \dots \quad (4.39b)$$

The expansion (4.38) is that for small  $\epsilon$  and  $\delta$  and, for the impurity, the values of eqs. (4.39a) and (4.39b) are correct so far as the first order of  $\epsilon$  and  $\delta$  is concerned. In Fig. 1 we have plotted  $\Delta F$  due to one isotope or hole. The deviations from the classical treatment of Kirkwood and Stripp<sup>1)</sup> are seen clearly.

Table 2.

$$f_1 = \frac{1}{2} (G_0 - 1) \zeta(2)$$

$$f_2 = -\frac{1}{8} (G_0 + 2) (G_0 - 1) \zeta(4)$$

$$f_3 = \frac{1}{48} (G_0 - 1) \{15 + 9(G_0 - 1) + 2(G_0 - 1)^2\} \zeta(6)$$

.....

where  $G_0$  is given by (4.35a).

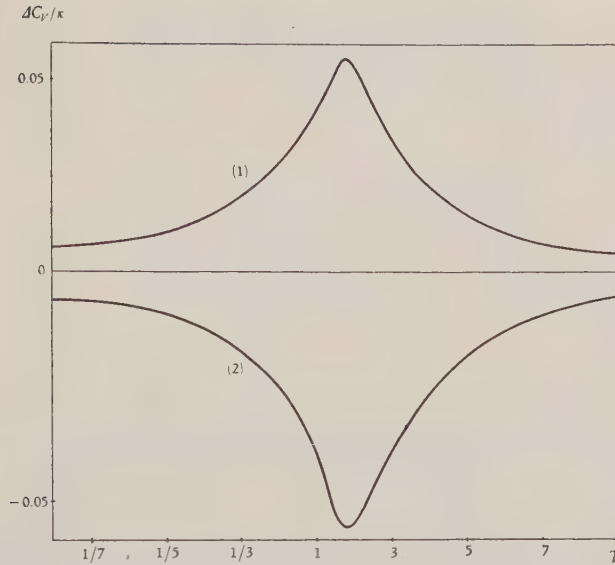


Fig. 2. Change of heat capacity due to one isotope or hole in the monatomic linear chain.

$$(1) \dots M' = (4/3)M \text{ or } \gamma' = (3/4)\gamma$$

$$(2) \dots M' = (2/3)M \text{ or } \gamma' = (3/2)\gamma.$$

Finally, the change of heat capacity due to one defect is obtained at once from (2.16). We have plotted the schematic graph of  $\Delta C_v$  in Fig. 2.

#### 4/3) Diatomic simple lattice.

It has already been shown in (I) that the equation of motion of the atom in the diatomic simple lattice reduces to that in the monatomic simple lattice by the following transformation:

$$\mathbf{v}_s = [\omega^2 \omega_L^2 \mu - 2 \sum_{k=1}^n \gamma_k]^{1/2} \mathbf{u}_s$$

$$\mu = \begin{cases} M_1 & \text{when } \sum_{i=1}^n s_i \text{ is even} \\ M_2 & \text{when } \sum_{i=1}^n s_i \text{ is odd,} \end{cases} \quad (4.41)$$

and

$$\omega^2 \omega_L^2 \mathbf{M}^* = 2 \sum_{k=1}^n \gamma_k + [(\omega^2 \omega_L^2 M_1 - 2 \sum \gamma_k)(\omega^2 \omega_L^2 M_2 - 2 \sum \gamma_k)]^{1/2} \quad (4.42)$$

Furthermore, the optical and acoustical bands are obtained by

$$\omega^2 \omega_L^2 \mathbf{M}^* = 2 \sum_{k=1}^n \gamma_k (1 - \cos \varphi_k). \quad (4.43)$$

In the case of linear chain, for instance, eq. (3.5) becomes



$$\begin{aligned} [\omega^2 \omega_L^2 M_1 - 2\gamma] u_{2s} + \gamma u_{2s+1} + \gamma u_{2s-1} &= 0 \\ [\omega^2 \omega_L^2 M_2 - 2\gamma] u_{2s+1} + \gamma u_{2s+2} + \gamma u_{2s} &= 0 \end{aligned} \quad (4.44)$$

which, by the transformation

$$\begin{aligned} v_{2s} &= [\omega^2 \omega_L^2 M_1 - 2\gamma]^{1/2} u_{2s} \\ v_{2s+1} &= [\omega^2 \omega_L^2 M_2 - 2\gamma]^{1/2} u_{2s+1}, \end{aligned}$$

reduces to the equation

$$[\omega^2 \omega_L^2 M^* - 2\gamma] v_s + \gamma v_{s+1} + \gamma v_{s-1} = 0 \quad (4.45)$$

where

$$\omega^2 \omega_L^2 M^* = 2\gamma + [(\omega^2 \omega_L^2 M_1 - 2\gamma)(\omega^2 \omega_L^2 M_2 - 2\gamma)]^{1/2}. \quad (4.46)$$

Eq. (4.45) reduces to the equation of motion of monatomic linear chain if  $M^*$  is replaced by  $M$ .

Then the Green function  $a^{-1}(s, s')$  can be calculated in the same way as that of monatomic lattice. The expression of  $z$ , however, becomes different due to the fact that  $M^*$  is the function of  $\omega$ . Then it can be easily proved that the Green function is given by (4.31) if  $z$  is replaced by

$$\begin{aligned} z = \frac{1}{2\gamma} \Big\{ & [(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma)]^{1/2} \\ & - [(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma) - 4\gamma^2]^{1/2} \Big\}. \end{aligned} \quad (4.47)$$

The characteristic functions for one defect are calculated in the usual way and the results are

$$\begin{aligned} \frac{D(i\omega'; \epsilon_1)}{D_0(i\omega'; \epsilon_1)} &= 1 - \epsilon_1 \omega'^2 \omega_L^2 M_1 \left[ \frac{(\omega'^2 \omega_L^2 M_2 + 2\gamma) / (\omega'^2 \omega_L^2 M_1 + 2\gamma)}{(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma) - 4\gamma^2} \right]^{1/2} \\ &\quad ; \text{ isotope,} \end{aligned} \quad (4.48a)$$

$$\begin{aligned} \frac{D(i\omega'; \delta)}{D_0(i\omega'; \delta)} &= 1 - \delta \left[ \frac{(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma) - 4\gamma^2}{(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma) - 4\gamma^2} \right]^{1/2} \\ &\quad \times [(\omega'^2 \omega_L^2 M_1 + 2\gamma)(\omega'^2 \omega_L^2 M_2 + 2\gamma)]^{-1/2} \\ &\quad ; \text{ hole,} \end{aligned} \quad (4.48b)$$

where

$$\epsilon_1 = 1 - \frac{M_2'}{M_1}, \quad \delta = 1 - \frac{\gamma}{\gamma'}.$$

Therefore,  $\Delta F$  is given by

$$\Delta F = \frac{1}{2\beta} \log G_0(\epsilon_{\frac{1}{2}}, \delta) + \frac{1}{\beta} \sum_{k=0}^{\infty} k \log G(\epsilon_{\frac{1}{2}}, \delta; (k+1)T) \quad (4.49)$$

with

$$G_0(\epsilon_{\frac{1}{2}}, \delta) = \begin{cases} (1 - \epsilon_{\frac{1}{2}})^{-1} & ; \text{ isotope,} \\ (1 - \delta)^{-1} & ; \text{ hole,} \\ (1 - \delta)^{-2} (1 - \epsilon_{\frac{1}{2}})^{-1} & ; \text{ impurity,} \end{cases} \quad (4.50a)$$

and

$$G_0(\epsilon_{\frac{1}{2}}, \delta; kT) = \begin{cases} 1 - \frac{\epsilon_{\frac{1}{2}}}{A(kT)} M_1 \omega_L^2 k^2 T^2 [\omega_L^2 M_1 k^2 T^2 + 2\gamma] & ; \text{ isotope,} \\ 1 - \frac{\delta}{A(kT)} [(\omega_L^2 M_1 k^2 T^2 + \gamma)(\omega_L^2 M_2 k^2 T^2 + \gamma) - \gamma^2] & ; \text{ hole,} \end{cases} \quad (4.50b)$$

where

$$\begin{aligned} A(kT) &= [(\omega_L^2 M_1 k^2 T^2 + 2\gamma)(\omega_L^2 M_2 k^2 T^2 + 2\gamma) - 4\gamma^2]^{1/2} \\ &\times [(\omega_L^2 M_1 k^2 T^2 + 2\gamma)(\omega_L^2 M_2 k^2 T^2 + 2\gamma)]^{1/2}, \\ \omega_L^2 &= \frac{1}{2\gamma} \left( \frac{1}{M_1} + \frac{1}{M_2} \right). \end{aligned} \quad (4.50c)$$

The generalization to the  $n$ -dimensional case is straightforward but tedious.

Comparing (4.50a) with (4.35a), it may be clear that the sign of  $\Delta F$  is controlled by the sign of  $\epsilon_{\frac{1}{2}}$  which in turn is controlled by the relative magnitude of  $M_1$  and  $M_2$ . Therefore, the defect whose mass is between  $M_1$  and  $M_2$  is apt to be replaced by the lighter atom. At low temperature, however, such a simple discussion may not be possible. Actually, the calculated expression of  $\Delta E_0$  has not such a simple form<sup>7)</sup>.

### § 5. Interaction energy of defect

We define the characteristic function owing to the correlation of  $m$  defects,  $K_1, K_2, \dots, K_m$ , as follows:

$$\begin{aligned} &G_m^I(\omega'; K_1, K_2, \dots, K_m) \\ &= \frac{G(\omega'; K_1 + K_2 + \dots + K_m)}{\prod_{j=1}^m G(\omega'; K_j)} \left[ 1 - \sum_{i=2}^{m-1} \sum_{(j_1, \dots, j_i)} G_i^I(\omega'; K_{j_1}, K_{j_2}, \dots, K_{j_i}) \right] \end{aligned} \quad (5.1)$$

where

$$G(\omega'; K_1 + K_2 + \cdots + K_m) = \frac{D(i\omega'; K_1 + K_2 + \cdots + K_m)}{D_0(i\omega'; K_1 + K_2 + \cdots + K_m)} \quad (5.2)$$

is the characteristic function when  $m$  defects are present. Summation with respect to  $(j_1, j_2, \dots, j_t)$  is taken over all the  $\binom{m}{t}$  sets. Especially, when  $m=2$ ,

$$G_2'(\omega'; K_1, K_2) = \frac{G(\omega'; K_1 + K_2)}{G(\omega'; K_1)G(\omega'; K_2)} - 1. \quad (5.1a)$$

From this definition the characteristic function of  $m$  defects, (5.2), is expanded as follows.

$$G(\omega'; K_1 + \cdots + K_m) = \prod_{j=1}^m G(\omega'; K_j) \left[ 1 + \sum_{t=2}^m \sum_{(j_1, \dots, j_t)} G_t'(\omega'; K_{j_1}, \dots, K_{j_t}) \right]. \quad (5.3)$$

Now, since the interaction energy is defined as

$$\Delta F'(K_1, \dots, K_m) = \Delta F(K_1 + K_2 + \cdots + K_m) - \sum_{j=1}^m \Delta F(K_j), \quad (5.4)$$

we get from (2.12), (5.1) and (5.3),

$$\Delta F'(K_1, K_2, \dots, K_m) = \sum_{t=2}^m \Delta F_t'(K_{j_1}, K_{j_2}, \dots, K_{j_t}) \quad (5.5)$$

with

$$\Delta F_t' = \frac{1}{2\beta} [A_t'(0) - A_t'(\infty)] + \frac{1}{\beta} \sum_{k=0}^{\infty} k [A_t'(kT) - A_t'((k+1)T)] \quad (5.6a)$$

and

$$A_t'(\omega') = \log \left[ 1 + \frac{\sum_{(j_1, \dots, j_t)} G_t'(\omega'; K_{j_1}, \dots, K_{j_t})}{1 + \sum_{p=2}^m \sum_{(j_1, \dots, j_p)} G_p'(\omega'; K_{j_1}, \dots, K_{j_p})} \right]. \quad (5.6b)$$

If we consider the isotopic defect, it is easily proved that the first term of (5.6a) vanishes. In the elements of the determinant  $G(K_1 + K_2 + \cdots + K_m; \omega')$  there exist those not included in the determinants  $G(K_i; \omega')$ 's,  $i=1, 2, \dots, m$ . Because those elements have the form that is proportional to  $\omega'^{-1} a^{-1}(\mathbf{s}, \mathbf{s}'; i\omega')$  where  $\mathbf{s} \neq \mathbf{s}'$ , they vanish at  $\omega'=0$  and  $\omega'=\infty$  from (4.9) and so the first term of (5.6a) is zero. This means that the interaction energy of defects is as small as can be regarded as the correlation to  $\Delta F$  of one defect and that vanishes at infinitely high temperatures, which may be expected physically. In fact this can also be proved in the case of the impurities of the linear chain. Then, it may be concluded that the first term of (5.6a) is zero in general and (5.6a) becomes

$$\Delta F_t' = \frac{1}{\beta} \sum_{k=0}^{\infty} k [A_t'(kT) - A_t'((k+1)T)]. \quad (5.7)$$

## 5/1): Interaction energy between two defects.

First, we investigate the correlation between two defects. In order to know the essential properties of interaction it is enough to consider only the isotopic case, although the impurity can be discussed in the same way.

In the monatomic linear chain the isotope and the hole are equivalent and we can treat them simultaneously. If we put

$$K = \begin{cases} \epsilon & \text{for isotope,} \\ \delta & \text{for hole,} \end{cases} \quad (5.8)$$

the characteristic function due to the correlation of two defects calculated from (5.1a) is given by

$$G_2'(\omega'; K_1, K_2) = \mp K_1 K_2 \frac{\omega'^2 [\sqrt{1+\omega'^2} - \omega']^{4|s_1-s_2| \mp 1 + 1}}{1 + \omega'^2} \frac{\left\{ 1 - K_1 \frac{\omega'}{\sqrt{1+\omega'^2}} \right\} \left\{ 1 - K_2 \frac{\omega'}{\sqrt{1+\omega'^2}} \right\}}{\left\{ 1 - K_1 \frac{\omega'}{\sqrt{1+\omega'^2}} \right\} \left\{ 1 - K_2 \frac{\omega'}{\sqrt{1+\omega'^2}} \right\}}, \quad (5.9)$$

where the minus sign is taken when the defects  $K_1, K_2$ , are of the same sort and the plus of the different sort. Inserting (5.9) into (5.7) we get the interaction energy of two defects,

$$\Delta F_2' = \sum_{k=1}^{\infty} \log[1 + G_2'(kT; K_1, K_2)], \quad (5.10)$$

where we have used the fact that the absolute magnitude of  $G_2'(K_1, K_2; \omega')$  is less than unity for all the values of  $K_1, K_2$  and  $\omega'$ . Also using this fact it is clear that the sign of  $\Delta F_2'$  is decided by the sign of  $G_2'(K_1, K_2; \omega')$ , that is, the sign of  $(\mp K_1 K_2)$ . In other words it is controlled by the sign of  $(\mp K_1 K_2)$  according to whether the interaction between two defects are attractive or repulsive. This is shown in Table 3. The result involves that derived in (I) as a special case.

Table 3.

	$\Delta F_2'$	$K_1$	$K_2$	
attractive	$(-)$	$M_1' < M(\gamma_1' > \gamma)$	$M_2' < M(\gamma_2' > \gamma)$	same sort
	$(-)$	$M_1' > M(\gamma_1' < \gamma)$	$M_2' > M(\gamma_2' < \gamma)$	
	$(-)$	$M_1' < M$	$\gamma_2' < \gamma$	different sort
	$(-)$	$M_1' > M$	$\gamma_2' > \gamma$	
repulsive	$(+)$	$M_1' < M(\gamma_1' > \gamma)$	$M_2' > M(\gamma_2' < \gamma)$	same sort
	$(+)$	$M_1' > M(\gamma_1' < \gamma)$	$M_2' < M(\gamma_2' > \gamma)$	
	$(+)$	$M_1' < M$	$\gamma_2' > \gamma$	different sort
	$(+)$	$M_1' > M$	$\gamma_2' < \gamma$	



Although the above discussion holds at higher temperatures, it is also valid at low temperatures. It is seen at once because the interaction part of  $JE_0$  is from (2.14) and (5.9),

$$JE_0' = \frac{\hbar\omega_L}{2\pi} \int_0^\infty \log[1 + G_2'(\omega'; K_1, K_2)] d\omega'. \quad (5.11)$$

Clearly the sign of  $JE_0$  is determined by that of the integrand of (5.11). This integral was calculated in (I) in the special case of small  $K_1$  and  $K_2$ . That is,

$$JE_0' \simeq (\mp K_1 K_2) \frac{\hbar\omega_L}{4\pi} \left\{ \frac{2(4s+1 \mp 1)}{16(4s+1 \mp 1)^2 - 1} - \psi\left(\frac{3}{4} + (4s+1 \mp 1)\right) + \psi\left(\frac{1}{4} + (4s+1 \mp 1)\right) \right\}; \quad s = |s_1 - s_2|,$$

while, at high temperature (5.10) becomes approximately

$$JE_2' \simeq \frac{1}{\beta} \log[1 \mp K_1 K_2 \zeta(4s) \left(\frac{1}{2T^2}\right)^{(4s+1 \mp 1)}]. \quad (5.12)$$

These results are at once extended to the case of isotopes of  $n$ -dimensional monatomic simple lattice. The correlational characteristic function of two isotopes  $\epsilon_s, \epsilon_{s'}$  is given by

$$G_2'(\omega'; \epsilon_s, \epsilon_{s'}) = \frac{(-\epsilon_s \epsilon_{s'}) [2n\omega'^2 P_n(s-s'; \omega')]}{\{1 - 2n\omega'^2 P_n(0 \cdots 0; \omega') \epsilon_s\} \{1 - 2n\omega'^2 P_n(0 \cdots 0; \omega_{st}) \epsilon_{s'}\}}. \quad (5.13)$$

where  $P_n(s-s'; \omega')$  is given in (4.5). Also in this case it can be proved  $|G_2'(K_s, K_{s'}; \omega')| < 1$  and, then, the discussion about the sign of  $JE_2'$  is retained.

The high and low temperature expansions can be done in the same way as in (4.1).

(ii) altogether. The results are:

$$JE_2' \simeq \frac{1}{\beta} \log[1 - \epsilon_s \epsilon_{s'} \zeta(4s) \left(\frac{1}{2nT^2}\right)^{4s}];$$

$$s = \sum_{j=1}^n |s_j - s_j'|, \quad (5.14)$$

$$JE_0' \simeq -\epsilon_s \epsilon_{s'} \frac{3\hbar\omega_L}{16\pi} \left(\frac{\pi}{2n}\right)^{1/2} p_{5/2}^{(2)}(s-s'); \quad$$

$$p_{5/2}^{(2)} = \text{const} = \int_0^\infty \int_0^\infty dx dx' \frac{e^{-n(x+xf)}}{(x+x')^{5/2}} \prod_{j,k=1}^n I_{s_j-s_j'}(x) I_{s_k-s_k'}(x'). \quad (5.15)$$

Eq. (5.15) is the result of small  $\epsilon_s$  and  $\epsilon_{s'}$ .

For a pair of isolated isotopes at a separation  $R = (\sum (s_j - s_j')^2)^{1/2} \gg 1$ , we find the following distance dependency of  $JE_I$  at high temperatures,

$$JE_I' \simeq [P_n(s-s'; T)]^2 \propto R^{-(n-1)} \exp[-M\omega_L^2 T^2 R],$$

while at low temperatures according to (I)

$$\Delta E_0^I \propto R^{-(2n+1)}.$$

Obviously the correlation of a pair of isotopes at high temperatures is more short-ranged than that at low temperatures. This result is to be compared with the result of Stripp and Kirkwood<sup>1)</sup> that the correlation of a pair of Schottky defects is proportional to  $R^{-6}$  at high temperatures.

Finally, we calculated the characteristic function due to the correlation of three or four isotopes. The results are

$$G_3^I(\omega'; K_1, K_2, K_3) = 2II_{\substack{\text{cycle} \\ (1,2,3)}} \frac{\epsilon_1 2n\omega'^2 P_n(\mathbf{s}_1 - \mathbf{s}_2; \omega')}{1 - \epsilon_1 2n\omega'^2 P_n(0 \cdots 0; \omega')}, \quad (5.16a)$$

$$G_4^I(\omega'; K_1, K_2, K_3, K_4) = \sum_{(r \neq i, j, k, l)} II_{(ijkl)} (\pm 1) \frac{\epsilon_i 2n\omega'^2 P_n(\mathbf{s}_i - \mathbf{s}_j; \omega')}{1 - \epsilon_i 2n\omega'^2 P_n(0 \cdots 0; \omega')}. \quad (5.16b)$$

The summation of the right-hand side of (5.16b) is extended over all the permutations of rings. The minus sign is taken when the ring is united, while the plus is taken when the ring of four defects are divided into two rings, one of which is composed of two defects. From this the interaction energy due to three or four defects can be calculated at once. The magnitudes of interactions are  $O(G_2^I(\omega'; K_1, K_2)^{3/2})$  and  $O(G_2^I(\omega'; K_1, K_2)^{4/2})$ , respectively. Remembering  $G_2^I(\omega'; K_1, K_2)^{1/2} < 1$ , it is clear that the strength of higher order interaction decreases as the order increases.

### 5/2) Interaction of a defect with the boundaries.

We have used the Born-von Kármán condition till now. However, if we use other boundary conditions, for instance, rigid or free boundary, there appears additional term in  $\Delta F$ , which we may interpret as the interaction term between the defects and the boundaries.

Understanding the essential features, it is enough to consider only the monatomic linear chain. Since the wave functions and the eigenvalues are determined so as to satisfy the boundary conditions, Green function  $a^{-1}(s, s')$  also depends upon them. Neglecting  $O(1/N)$ , we obtain them by simple calculation. (See (I).)

$$a^{-1}(s, s'; i\omega') = \begin{cases} \frac{1}{\gamma} \frac{z^{s-s'} - z^{-s-s'}}{z - z^{-1}} & ; B.K. \\ \frac{1}{\gamma} \frac{z^{|s-s'|} - z^{|s+s'|}}{z - z^{-1}} & ; R.B. \\ \frac{1}{\gamma} \frac{z^{|s-s'|} + z^{|s+s'-1|}}{z - z^{-1}} & ; F.B. \end{cases} \quad (5.17)$$

$$z = [v'1 + \omega'^2 - \omega']^2.$$

Now, we define the interaction energy of a defect with the boundaries as

$$\Delta F_{F.B.}^I = \Delta F_{R.B.}^I - \Delta F_{B.K.}^I. \quad (5.18)$$

If the defect is an isotope or a hole, from the simple calculation like Appendix II eq. (5.18) becomes

$$\Delta F_{F.B.}^I = \sum_{k=1}^{\infty} \log[1 + G_{R.B.}^I(kT; K')] \quad (5.19)$$

with

$$G_{F.B.}^I = \pm K' \frac{\omega'}{\sqrt{1+\omega'^2}} \frac{[\sqrt{1+\omega'^2} - \omega']^{2s-1 \pm 1}}{1 - K' \frac{\omega'}{\sqrt{1+\omega'^2}}} \quad (5.19a)$$

where  $s$  is the distance between the defect and the boundary. Again, since  $G^I(K'; \omega') < 1$ ,

$$(\Delta F_{F.B.}^I) \geq 0 \quad \text{when } (\pm K') \geq 0,$$

or

$$\begin{aligned} \text{attractive } (\Delta F^I < 0) \dots & \begin{cases} M' < M & (\gamma' > \gamma) \dots R.B. \\ M' > M & (\gamma' < \gamma) \dots F.B. \end{cases} \\ \text{repulsive } (\Delta F^I > 0) \dots & \begin{cases} M' > M & (\gamma' < \gamma) \dots R.B. \\ M' < M & (\gamma' > \gamma) \dots F.B. \end{cases} \end{aligned}$$

The high and low temperature expansions are given by usual calculations; namely,

$$\Delta F_{F.B.}^I \simeq \frac{1}{\beta} \log[1 \pm K' \zeta(2s - \frac{1}{2} \pm \frac{1}{2}) \left( \frac{1}{2T^2} \right)^{2s-1/2 \pm 1/2}], \quad (5.20)$$

$$\Delta E_{F.B.}^I \simeq \pm K' \frac{\hbar \omega_L}{32\pi} - \frac{1}{s^2} \quad (K' \sim 0). \quad (5.21)$$

Eq. (5.21) has already been obtained in (I) for the case of isotope.

## § 6. Free energy of the lattice with irregular distributed defects

We shall interpret the lattice with irregularly distributed defects as such that each atom of the perfect lattice is accidentally replaced by a defect with a certain probability. Then the characteristic functions corresponding to such lattices are obtained by following two processes; (a) fixing the total number of defects and taking an average with respect to all the possible relative configurations of defects and then, (b) taking an average with respect to all the possible total numbers of defects.

When the total number of defects  $m$  is of the order  $N$ , the characteristic determinant becomes that of the order  $N$ , which is, of course, impossible to calculate directly. As was noticed at the end of §5.1, however, the higher order correlations may be neglected if the correlation of defects is not so strong. Therefore, we have only to calculate the first several terms of (5.3) and may put approximately

$$\langle G(\omega'; K_1 + K_2 + \dots + K_m) \rangle \simeq \prod_{j=1}^m G(\omega'; K_j) \exp \sum_t \left[ \left\langle \sum_{(j_1 \dots j_t)} G_t'(\omega'; K_{j_1}, \dots, K_{j_t}) \right\rangle \right]. \quad (6.1)$$

where the notation  $\langle \rangle$  means the average of type (a). Since the function  $G(\omega'; K_j)$  is independent of the relative configurations of defects it is invariant by the average of type (a).

From (2.12), (5.2) and (6.1) we obtain

$$\langle \mathcal{J}F(K_1 + K_2 + \dots + K_m) \rangle = \sum_{j=1}^m \mathcal{J}F(K_j) + \langle \mathcal{J}F^I(K_1, K_2, \dots, K_m) \rangle \quad (6.2)$$

where

$$\langle \mathcal{J}F^I(K_1, K_2, \dots, K_m) \rangle \simeq \sum_{k=0}^{\infty} \sum_t \left\langle \sum_{(j_1 \dots j_t)} G_t'(kT; K_{j_1}, \dots, K_{j_t}) \right\rangle. \quad (6.3)$$

In (6.3) we have rearranged the series with respect to  $k$ .

Henceforth, we shall treat the monatomic linear chain with the isotopic defects (or holes). Our results obtained here can be easily extended to the simple cubic lattice and the impurity, although the calculation becomes tedious. Also we shall consider, for simplicity, all the  $K_j$ 's to be equal to  $K$ .

Now, we shall take the average of type (a). First, we suppose that the probability, with which each normal atom is replaced by a defect, is independent on the configuration of other defects; namely, it is determined only by the concentration of defects  $m/N$ . Then, remembering  $G_2^I(\omega'; K_j, K_k)$  is given by (5.9) and

$$\begin{aligned} \left\langle \sum_{(j,k)} z^{2|s_j - s_k|} \right\rangle &= \frac{1}{2} \sum_{s_j=1}^N \left( \frac{m}{N} \right) \left( \sum_{s=1}^{s_j-1} + \sum_{s=1}^{N-s_j} \right) \left( \frac{m}{N} \right) z^{2s} \\ &= m \left( \frac{m}{N} \right) \left[ \frac{z^2}{1-z^2} + O\left(\frac{1}{N}\right) \right] \end{aligned}$$

we get the average value of (5.9),

$$\begin{aligned} &\left\langle \sum_{(j,k)} G_2^I(\omega'; K_j, K_k) \right\rangle \\ &= m \left( \frac{m}{N} \right) (-K^2) \frac{\omega'^2 / (1 + \omega'^2)}{\left\{ 1 - K \frac{\omega'}{\sqrt{1 + \omega'^2}} \right\}} \frac{z^{2+1/2 \mp 1/2}}{1 - z^2}. \end{aligned} \quad (6.4)$$

Substituting (6.4) in (6.2) and (6.3), we obtain



$$\langle \Delta F(mK) \rangle = m[\Delta F(K) + \frac{1}{m} \langle \Delta F^I(mK) \rangle] \quad (6.5)$$

where  $(1/m) \langle \Delta F^I(mK) \rangle$  is the mean interaction energy per pair of defects and is given by

$$\begin{aligned} & \langle \frac{1}{m} \Delta F^I(mK) \rangle \\ &= \binom{m}{N} (-K^2) \sum_{k=1}^{\infty} \frac{k^2 T^2}{1 + k^2 T^2} \left[ 1 - K \frac{kT}{1 + k^2 T^2} \right] \left[ 1 - \frac{1}{1 + k^2 T^2} - kT \right] \quad (6.6) \end{aligned}$$

Eq. (6.6) is the first term of the expansion of interaction energy with respect to concentration. The next terms are obtained by calculating the terms  $t=2, 3, \dots$  in the right-hand side of (6.3). It may be clear that the interaction energy per triangle of defects is  $O(m^2/N^2)$  which is small compared with that of pair, when the concentration  $m/N$  is much smaller than unity; because the pair's is of the order  $O(m/N)$ .

Next, we suppose that the probability, with which a normal atom at a distance  $s$  from a defect is replaced by a defect, is proportional to  $\exp(-\Delta F(s)/kT)$ . This assumption seems to take account of the correlations between defects approximately. Then

$$\begin{aligned} \langle \sum z^{2|s_j - s_k|} \rangle &= -N \binom{m}{N}^2 \frac{\partial}{\partial f} \log \left[ \sum_{s_j=1}^N \left( \sum_{s=1}^{s_j-1} + \sum_{s=1}^{N-s_j} \right) \exp(-fz^{2s}) \right] \\ &= m \binom{m}{N} \sum_{q=0}^{\infty} \frac{(-f)^q}{q!} \frac{z^{2q}}{(1 - z^{2q+2})} \left[ \sum_{q'=0}^{\infty} \frac{(-f)^{q'}}{q'!} \frac{z^{2q'}}{1 - z^{2q'}} \right] \quad (6.7) \end{aligned}$$

where  $f$  is determined by the condition:

$$\frac{1}{N} \sum_{s_j=1}^N \left( \sum_{s=1}^{s_j-1} + \sum_{s=1}^{N-s_j} \right) \exp(-fz^{2s}) = \frac{m}{N}.$$

The summation (6.7) may not be expressed in the closed form, but it is sure that the value of (6.7) is between  $m(m/N)z^2$  and  $m(m/N)z^2/(1-z^2)$  and may nearly equal to  $m(m/N)z^2$ . Hence the average value of (5.9) becomes in the present case

$$\frac{1}{m} \langle \Delta F^I(mK) \rangle \simeq \binom{m}{N} (-K^2) \sum_{k=1}^{\infty} \frac{k^2 T^2}{\sqrt{1 + k^2 T^2}} \left[ 1 - K \frac{kT}{\sqrt{1 + k^2 T^2}} \right] \left[ 1 - \frac{1}{\sqrt{1 + k^2 T^2}} - kT \right] \quad (6.8)$$

which has to be replaced for (6.6). Clearly, if the other factors are not considered, (6.7) may be more probable than (6.6).

We shall, then consider to take the average of type (b). If the mean concentration

$m_0/N$  is much less than unity, we may approximate the binary distribution to the Poisson's. Then,

$$\begin{aligned}\overline{\langle G(\omega'; m_0 K) \rangle} &= e^{-m_0} \sum_{m=0}^N \frac{(m_0)^m}{m!} [G(\omega'; K)]^m \exp \left[ \sum_t \left\langle \sum_{j_1 \dots j_t} G_t^j(\omega'; tK) \right\rangle \right] \\ &\cong \exp[m_0 G(\omega'; K)] \exp \left\{ \frac{1}{m_0} \left\langle \sum_{(j,k)} G_2^j(\omega') \right\rangle_{m=m_0} - m_0 \right\}\end{aligned}\quad (6.9)$$

where we have replaced  $(1/m) \langle \sum G_2^j(\omega') \rangle$  by  $(1/m_0) \langle \sum G_2^j(\omega') \rangle_{m=m_0}$  because it mainly contributes to the average where  $m \cong m_0$  and also because  $\exp(m_0^{-1} \langle \sum G_2^j \rangle)$  is very nearly equal to unity. The notation "bar" means the average of type (b). Use of (6.8) and (2.12) leads at once to

$$\overline{\langle \Delta F(m_0 K) \rangle} = m_0 [\overline{\Delta F(K)} + \overline{\langle \Delta F'(m_0 K) \rangle}] \quad (6.10)$$

with

$$\overline{\Delta F(K)} = \frac{K}{2\beta} + \frac{1}{\beta} \sum_{k=0}^{\infty} (-K) k \left\{ \frac{kT}{v' 1 + k^2 T^2} - \frac{(k+1)T}{v' 1 + (k+1)^2 T^2} \right\} \quad (6.11)$$

and

$$\begin{aligned}\overline{\langle \Delta F'(m_0 K) \rangle} &= \frac{1}{\beta} \sum_{k=0}^{\infty} k [G(kT; K) \{ \exp(\langle \sum_{m_0} \frac{1}{m_0} G_2^j(kT; m_0 K) \rangle) - 1 \} \\ &\quad - G(kT + T; K) \{ \exp(\langle \sum_{m_0} \frac{1}{m_0} G_2^j(kT + T; m_0 K) \rangle) - 1 \}],\end{aligned}\quad (6.12)$$

where  $G(\omega'; K)$  is given by (4.32) and  $(1/m_0) \langle \sum G_2^j(\omega'; m_0 K) \rangle$  is obtained by putting  $m=m_0$  in (6.4) or (6.8).

The high and low temperature expansions are also easily calculated. The results are

$$\overline{\langle \Delta F(m_0 K) \rangle} = \frac{1}{\beta} m_0 K \left[ \frac{1}{2} - \sum_{l=1}^{\infty} \left( -\frac{1}{l^2} \right) \zeta(2l) \left( \frac{1}{T} \right)^{2l} \right] + \overline{\langle \Delta F'(m_0 K) \rangle} \quad (6.13)$$

and

$$\overline{\langle \Delta E_0(m_0 K) \rangle} = \frac{\hbar \omega_L}{2\pi} m_0 K + \overline{\langle \Delta E_0'(m_0 K) \rangle} \quad (6.14)$$

where we have omitted the detailed expressions of  $\overline{\langle \Delta F'(m_0 K) \rangle}$  and  $\overline{\langle \Delta E_0'(m_0 K) \rangle}$  because they are of rather complicated forms.

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## Appendix I

The characteristic function of the simple cubic lattice due to one impurity is the seventh order determinants given by

$$\frac{D(i\omega')}{D_0(i\omega')} = \begin{vmatrix} 1 + \rho'_1 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_3 J_3 \\ -\rho'_2 J_2 & 1 + \rho'_1 J_2 & -\rho'_2 J_2 & -\rho'_2 J_2 & -\rho'_2 J_2 & -\rho'_3 J_2 & -\rho'_2 J_2 \\ -\rho'_2 J_1 & -\rho'_2 J_1 & 1 + \rho'_1 J_1 & -\rho'_2 J_1 & -\rho'_3 J_1 & -\rho'_2 J_1 & -\rho'_2 J_1 \\ J_3 & J_2 & J_1 & 1 + \rho'_1 J_1 & J_1 & J_2 & J_3 \\ -\rho'_2 J_1 & -\rho'_2 J_1 & -\rho'_3 J_1 & -\rho'_2 J_1 & 1 + \rho'_1 J_1 & -\rho'_2 J_1 & -\rho'_2 J_1 \\ -\rho'_2 J_2 & -\rho'_3 J_2 & -\rho'_2 J_2 & -\rho'_2 J_2 & -\rho'_2 J_2 & 1 + \rho'_1 J_2 & -\rho'_2 J_2 \\ -\rho'_3 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & -\rho'_2 J_3 & 1 + \rho'_1 J_3 \end{vmatrix} \quad (\text{A} \cdot 1)$$

with

$$\rho'_1 = a^{-1} \begin{pmatrix} 0 & 0 & 0 \end{pmatrix} - a^{-1} \begin{pmatrix} 0 & 1 & 1 \end{pmatrix}$$

$$\rho'_2 = a^{-1} \begin{pmatrix} 0 & 0 & 1 \end{pmatrix} - a^{-1} \begin{pmatrix} 0 & 1 & 1 \end{pmatrix}$$

$$\rho'_3 = a^{-1} \begin{pmatrix} 0 & 0 & 1 \end{pmatrix} - a^{-1} \begin{pmatrix} 0 & 0 & 2 \end{pmatrix}$$

where

$$a^{-1} \begin{pmatrix} 0 & 0 & 1 \end{pmatrix} = a^{-1} (\mathbf{s}_1 \ \mathbf{s}_2) \quad \text{with } \mathbf{s}_1 - \mathbf{s}_2 = \mathbf{s} \begin{pmatrix} 0 & 0 & 1 \end{pmatrix}$$

etc.,

and

$$J_i = \gamma_i \delta_i' = \gamma_i \left( 1 - \frac{\gamma_i'}{\gamma_i} \right) \quad (i=1, 2, 3)$$

$$J_j = M \omega'^2 \omega_L'^2 \in a^{-1} (\mathbf{s}, \mathbf{s}) + 2 (J_k + J_l) \rho'_2' + 2 J_j (\rho'_3' - \rho'_2')$$

$$(j, k, l=1, 2, 3; \text{ cyclic})$$

$$J' = M \omega'^2 \omega_L'^2 \in a^{-1} (\mathbf{s}, \mathbf{s}') + 2 (J_1 + J_2 + J_3) (\rho'_1' - \rho'_2').$$

If we consider the isotropic case  $\gamma_1 = \gamma_2 = \gamma_3 = \gamma$ ,  $\rho'_i$ 's ( $i=1, 2, 3$ ) reduce to  $-\gamma \rho'_i$ 's given by (4.8) and all the  $J_i$ 's and  $J_i$ 's become the same respectively. Hence (A.1) can be readily calculated and the result is (4.6).

## Appendix II

In the monatomic linear chain the equations of motion (4.1) become

$$\begin{aligned}
[\omega^2 \omega_L^2 M - (\gamma + \gamma')] u_{s-1} + \gamma' u_s + \gamma u_{s-2} &= 0 \\
[\omega^2 \omega_L^2 M - 2\gamma'] u_s + \gamma' u_{s+1} + \gamma u_{s-1} &= 0 \\
[\omega^2 \omega_L^2 M - (\gamma + \gamma')] u_{s+1} + \gamma u_{s+2} + \gamma' u_s &= 0.
\end{aligned} \tag{A.2}$$

Hence  $\mathcal{A}(s, s')$ 's of (3.6) are given by

$$\begin{aligned}
\mathcal{A}(s-1, s-1) &= \mathcal{A}(s+1, s+1) = \gamma \delta \equiv \mathcal{A}_2 \\
\mathcal{A}(s-1, s) &= \mathcal{A}(s, s-1) = \mathcal{A}(s, s+1) = \mathcal{A}(s+1, s) = -\mathcal{A}_2 \\
\mathcal{A}(s, s) &= M \omega'^2 \omega_L^2 \in a^{-1}(s, s) + 2\gamma \delta \equiv \mathcal{A}_1 + 2\mathcal{A}_2 \\
\mathcal{A}(s, s') &= 0 \quad \text{otherwise,}
\end{aligned} \tag{A.3}$$

and the characteristic determinants are

$$\begin{aligned}
&\frac{D(i\omega')}{D_0(i\omega')} = \\
&1 + \sum_{k=s-2}^s \mathcal{A}(s-1, k) a^{-1}(s-1, k), \sum_{k=s-2}^s \mathcal{A}(s-1, k) a^{-1}(s, k), \sum_{k=s-2}^s \mathcal{A}(s-1, k) a^{-1}(s+1, k) \\
&\sum_{k=s-1}^{s+1} \mathcal{A}(s, k) a^{-1}(s-1, k), 1 + \sum_{k=s-1}^{s+1} \mathcal{A}(s, k) a^{-1}(s, k), \sum_{k=s-1}^{s+1} \mathcal{A}(s, k) a^{-1}(s+1, k) \\
&\sum_{k=s}^{s+2} \mathcal{A}(s+1, k) a^{-1}(s-1, k), \sum_{k=s}^{s+2} \mathcal{A}(s+1, k) a^{-1}(s, k), 1 + \sum_{k=s}^{s+2} \mathcal{A}(s+1, k) a^{-1}(s+1, k)
\end{aligned} \tag{A.4}$$

Inserting (A.3) and (4.31) into (A.4) and after some calculations we get the following result :

$$\begin{aligned}
\frac{D(i\omega')}{D_0(i\omega')} &= 1 - (\epsilon + 2\delta) \frac{\omega'}{\sqrt{1 + \omega'^2}} + 4\delta\epsilon \frac{\omega'^2}{\sqrt{1 + \omega'^2}} [\sqrt{1 + \omega'^2} - \omega'] \\
&+ 4\delta^2 (1 - \epsilon) \frac{\omega'^3}{\sqrt{1 + \omega'^2}} [\sqrt{1 + \omega'^2} - \omega']^2.
\end{aligned} \tag{A.5}$$

This is the same as (4.32c). The characteristic functions due to one isotope is obtained at once by putting  $\delta=0$ , ( $\gamma'=\gamma$ ), in (A.5), and the one due to one hole is also obtained in a similar way.

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## Composite Model of Pion and Pion-Nucleon Interaction

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Regarding the pion as quantum of the lowest exciting mode of nucleon-antinucleon pair from the vacuum, we get a normalized wave function for the pion and use this wave function as an auxiliary variable for the treatment of the many body problem. Then we perform the canonical transformation and obtain an effective pion-nucleon interaction. The result is equivalent to the customary meson theory.

### § 1. Introduction

Various phenomena related with the low-energy  $p$ -wave pion-nucleon interaction were explained successfully by Chew and Low<sup>1)</sup> by using the cut-off Yukawa theory without nucleon recoil. But problems which seem to have a close connection with the  $s$ -wave pion-nucleon interaction, e. g.,  $s$ -wave pion-nucleon scattering, anomalous magnetic moment of the nucleon, the radius of the nucleon core, the electron-neutron interaction, etc., are still unsolved on the basis of a satisfactory theory. Probably, the virtual nucleon-antinucleon pair plays an important part in these problems. If an adequate method to treat the nucleon pair is known, these problems will be solved successfully.

On the other hand, in the composite model of the pion proposed by Fermi and Yang,<sup>2)</sup> the pion is regarded as a bound state of a nucleon and an antinucleon and, therefore, nucleon pairs play the essential role.\* Already Machida<sup>3)</sup> attempted to treat the pion-nucleon scattering on this model. We also here regard the pion as quantum of the lowest exciting mode of nucleons and antinucleons from the vacuum. In section 2 we consider the eigenvalue problem for the bound state of nucleon pairs using the new Tamm-Dancoff method, and identify this state with the pion. Then we obtain a normalized wave function for this state and use this wave function as an auxiliary variable for the treatment of the many body problem as in the case of plasma oscillation.<sup>4)</sup> In section 3 we perform the canonical transformation and obtain an effective pion-nucleon interaction.

We proceed along the treatment of the plasma oscillation formally, but, of course, there is a sharp difference physically. In the plasma oscillation repulsive, long range force plays the essential role, while strong, attractive, short range forces are concerned with our problem. Nevertheless the point that makes our treatment possible is that interactions between particle-antiparticle pairs play the main role.

\* A composite model of this sort was considered for strange particles by many authors.<sup>2)</sup>

## § 2. Eigenvalue problem for bound state of nucleon-antinucleon pair

We assume that the interaction Hamiltonian between nucleons and antinucleons has the following Fermi type,

$$H' = \frac{1}{4} \frac{g}{M^2} \sum_{i=1}^3 \int (\bar{\psi}^i(\mathbf{r}) \gamma_5 \tau_i \psi^i(\mathbf{r}) - \bar{\psi}^i(\mathbf{r}) \gamma_5 \tau_i \psi^i(\mathbf{r})) \times (\bar{\psi}^i(\mathbf{r}) \gamma_5 \tau_i \psi^i(\mathbf{r}) - \bar{\psi}^i(\mathbf{r}) \gamma_5 \tau_i \psi^i(\mathbf{r})) d\mathbf{r}, \quad (1)$$

where

$$\psi = \begin{pmatrix} \psi_p \\ \psi_n \end{pmatrix} \quad \text{and} \quad \psi^i = i \tau_2 C \bar{\psi} = \begin{pmatrix} \psi_n^i \\ -\psi_p^i \end{pmatrix} \quad (2)$$

are the nucleon and antinucleon field operators respectively.  $\tau_i$ 's are the usual isotopic spin operators. We assume the above interaction Hamiltonian because it is the simplest one which produces a bound state of the nucleon-antinucleon pair corresponding to the pseudoscalar pion. Of course this point interaction makes our treatment impossible as it stands, therefore, following the usual treatment of the pair theory,<sup>(4)</sup> we shall resort to a cut-off in momentum space chosen of the order of magnitude of the nucleon mass. We shall not write this cut-off function explicitly, but use this when necessary. The binding energy of the pion is about  $2M$  ( $M$  is the nucleon mass), so that the relative momenta of the nucleon and the antinucleon in the bound state are expected to extend to  $2M$ . However, higher cut-off momentum makes our mathematical treatment more difficult. Hence we consider here a fictitious model and cut off the relative momentum at the nucleon mass.

Now we represent the interaction Hamiltonian in terms of the creation and annihilation operators and pick up the terms which contribute to the nucleon-antinucleon interaction predominantly, neglecting higher powers of  $p/M$ .

Then we have

$$H' = \frac{1}{(2\pi)^3} \sum_{i, \rho} \frac{g}{M^2} \int \{ -a_{\mathbf{p}'+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^i b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_2*} + b_{-\mathbf{p}'+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_2*} \} \times \{ -a_{\mathbf{p}''-\mathbf{k}'/2}^{\rho_3*} B_{\rho_3 \rho_4}^i b_{-\mathbf{p}''+\mathbf{k}'/2}^{\rho_4*} + b_{-\mathbf{p}''-\mathbf{k}'/2}^{\rho_3} C_{\rho_3 \rho_4}^i a_{\mathbf{p}''+\mathbf{k}'/2}^{\rho_4*} \} d\mathbf{p}' d\mathbf{p}'' d\mathbf{k}', \quad (3)$$

where

$$B^i = (\tau_i \tau_2) \sigma_2 \\ C^i = (\tau_2 \tau_i) \sigma_2 \quad (4)$$

and  $a_p^*$  and  $b_p^*$  are creation operators and  $a_p$  and  $b_p$  annihilation operators for the nucleon and antinucleon respectively.

Next we consider the state which has one more nucleon-antinucleon pair than the true vacuum and treat this state by the use of the new Tamm-Dancoff method. For

this purpose we calculate the commutator between Hamiltonian and  $-a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1^*} B_{\rho_1 \rho_2}^j b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*}$  or  $b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1}$ . If we neglect the excited states which have two or more pairs compared with the vacuum, and neglect nucleon or antinucleon exchange terms between two pairs because it has smaller spin and isospin statistical weight than the non-exchange term, then we have the following commutators,

$$\begin{aligned} & [-a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1^*} B_{\rho_1 \rho_2}^j b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*}, H'] \\ &= \frac{8}{(2\pi)^3} \sum_{\rho_3 \rho_4} \frac{g}{M^2} \int d\mathbf{p}' \{ -a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_3^*} B_{\rho_3 \rho_4}^j b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_4^*} - b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_3^*} C_{\rho_3 \rho_4}^j a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_4} \}, \end{aligned} \quad (5)$$

$$\begin{aligned} & [b_{-\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}, H'] \\ &= -\frac{8}{(2\pi)^3} \sum_{\rho_3 \rho_4} \frac{g}{M^2} \int d\mathbf{p}' \{ -a_{\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} B_{\rho_3 \rho_4}^j b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_4^*} + b_{-\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} C_{\rho_3 \rho_4}^j a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_4} \}. \end{aligned} \quad (6)$$

In order to obtain the eigenvalue equation for the one pair excited state, we set up equations of motion for the operators  $-a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1^*} B_{\rho_1 \rho_2}^j b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*}$  and  $b_{-\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}$  in the Schroedinger representation. If we denote an eigenstate of the total Hamiltonian  $H(=H_0+H')$  corresponding to eigenvalue  $E$  by  $|T\rangle$  and the vacuum state which has the lowest eigenvalue  $E_0$  by  $|T_0\rangle$ , we have the following equations with the aid of the commutation relations (5) and (6),

$$\begin{aligned} & (|T\rangle, (H-E) (-a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1^*} B_{\rho_1 \rho_2}^j b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*}) |T_0\rangle) \\ &= (E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - E + E_0) (|T\rangle, -a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1^*} B_{\rho_1 \rho_2}^j b_{-\mathbf{p}-\mathbf{k}/2}^{\rho_2^*} |T_0\rangle) \\ &= \frac{8}{(2\pi)^3} \sum_{\rho_3 \rho_4} \frac{g}{M^2} \int d\mathbf{p}' (|T\rangle, (-a_{\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} B_{\rho_3 \rho_4}^j b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_4^*} + b_{-\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} C_{\rho_3 \rho_4}^j a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_4}) |T_0\rangle) \\ &= 0, \end{aligned} \quad (7)$$

$$\begin{aligned} & (|T\rangle, (H-E) b_{-\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2} |T_0\rangle) \\ &= (-E_{\mathbf{p}+\mathbf{k}/2} - E_{\mathbf{p}-\mathbf{k}/2} - E + E_0) (|T\rangle, b_{-\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2} |T_0\rangle) \\ &+ \frac{8}{(2\pi)^3} \sum_{\rho_3 \rho_4} \frac{g}{M^2} \int d\mathbf{p}' (|T\rangle, (-a_{\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} B_{\rho_3 \rho_4}^j b_{-\mathbf{p}'-\mathbf{k}/2}^{\rho_4^*} + b_{-\mathbf{p}'+\mathbf{k}/2}^{\rho_3^*} C_{\rho_3 \rho_4}^j a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_4}) |T_0\rangle) \\ &= 0. \end{aligned} \quad (8)$$

From these formulae we get the eigenvalue equation

$$\left\{1 - \frac{8}{(2\pi)^3} \frac{g}{M^2} \int d\mathbf{p} \left( \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}} \right) \right\} \\ \times \sum_{\rho_1 \rho_2} \int d\mathbf{p}' (\Psi, (-a_{\mathbf{p}'+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}'-\mathbf{k}/2}^{\rho_2*} + b_{\mathbf{p}'+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}'-\mathbf{k}/2}^{\rho_2}) \Psi_0) = 0, \quad (9)$$

that is

$$1 = \frac{8}{(2\pi)^3} \frac{g}{M^2} \int d\mathbf{p} \left( \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}} \right), \quad (10)$$

where

$$E - E_0 = \omega_{\mathbf{k}}.$$

This eigenvalue equation has two sorts of solutions. One is

$$\omega_{\mathbf{k}} = E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2}, \quad (11)$$

which corresponds to a free nucleon-antinucleon pair and the other is

$$\omega_{\mathbf{k}} \sim \sqrt{\mathbf{k}^2 + \mu^2}, \quad (12)$$

which corresponds to a bound state. The latter is obtained by integrating (10) and solving the resulting equation with respect to  $\omega_{\mathbf{k}}$ . In doing so we neglect terms higher than quadratic with respect to  $\mathbf{k}/M$  in square root of (12). If we cut the integral in (10) at the nucleon mass and take  $g=6.6$ , we can adjust  $\mu$  to the pion mass.

We regard this nucleon-antinucleon pair in the bound state as  $\pi$ -meson, and replace the fermion pair by a boson field effectively. We seek the explicit form of the wave function representing this bound state. This may be put in the following form,

$$\Psi_{\pi}^i(\mathbf{k}) = A_{\mathbf{k}}^{i*} \Psi_0, \quad (13)$$

$$A_{\mathbf{k}}^{i*} = \sum_{\rho_1 \rho_2} \int (-\alpha_{\mathbf{p}} a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^i b_{\mathbf{p}-\mathbf{k}/2}^{\rho_2*} + \beta_{\mathbf{p}} b_{\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}) d\mathbf{p}, \quad (14)$$

where  $\alpha_{\mathbf{p}}$  and  $\beta_{\mathbf{p}}$  are chosen so that  $\Psi_{\pi}^i(\mathbf{k})$  becomes a properly normalized eigenfunction of the total Hamiltonian. That is, using the equations

$$(H_0 + H') \Psi_{\pi}^i(\mathbf{k}) = (E_0 + \omega_{\mathbf{k}}) \Psi_{\pi}^i(\mathbf{k}), \quad (15)$$

$$(H_0 + H') \Psi_0 = E_0 \Psi_0 \quad (16)$$

and the commutation relations (5) and (6), we can see that  $\alpha_{\mathbf{p}}$  and  $\beta_{\mathbf{p}}$  are written as

$$\alpha_{\mathbf{p}} = \frac{-N_{\mathbf{k}}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} \\ \beta_{\mathbf{p}} = \frac{N_{\mathbf{k}}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}}, \quad (17)$$

where  $N_{\mathbf{k}}$  is determined from the normalization condition. Namely we can proceed as follows. (15) can also be written as an operator equation



$$[(H_0 + H'), A_k^{i*}] = \omega_k A_k^{i*}. \quad (18)$$

Taking the Hermite conjugate of the above equation,

$$[(H_0 + H'), A_k^i] = -\omega_k A_k^i. \quad (19)$$

From this we have

$$(H_0 + H') A_k^i \Psi_0 = (E_0 - \omega_k) A_k^i \Psi_0. \quad (20)$$

Since  $\Psi_0$  is the state of the lowest energy, this equation can be satisfied only if

$$A_k^i \Psi_0 = 0. \quad (21)$$

Using (13), (14) and this condition, we can fix the normalization as

$$\begin{aligned} (\Psi_\pi^i(\mathbf{k}), \Psi_\pi^j(\mathbf{k}')) &= (\Psi_0, A_k^i A_{k'}^{j*} \Psi_0) \\ &= (\Psi_0, [A_k^i, A_{k'}^{j*}] \Psi_0) \\ &= 4\delta_{ij} \delta(\mathbf{k} - \mathbf{k}') \int (|\alpha_p|^2 - |\beta_p|^2) d\mathbf{p} \\ &= \delta_{ij} \delta(\mathbf{k} - \mathbf{k}'), \end{aligned}$$

where we make the same approximation that leads to commutation relations (5) and (6). Thus the normalization condition is

$$4 \int d\mathbf{p} (|\alpha_p|^2 - |\beta_p|^2) = 1. \quad (22)$$

If we determine  $N_k$  from (22) and (17) using the cut-off procedure at the nucleon mass, (14) and its Hermite conjugate can be written approximately as

$$A_k^{i*} \simeq \frac{1}{\sqrt{2\omega_k}} \sum_{\rho_1 \rho_2} \int d\mathbf{p} \left( \frac{a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^i b_{\mathbf{p}-\mathbf{k}/2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} + \frac{b_{\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \right), \quad (23)$$

$$A_k^i \simeq \frac{1}{\sqrt{2\omega_k}} \sum_{\rho_1 \rho_2} \int d\mathbf{p} \left( \frac{a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1} B_{\rho_1 \rho_2}^i b_{\mathbf{p}+\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} + \frac{b_{\mathbf{p}-\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}+\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right). \quad (24)$$

### § 3. Effective Hamiltonian

Making use of the pion wave function obtained in the preceding section, we seek for an effective Hamiltonian for the pion-nucleon system. To this end, on the total Hamiltonian

$$H = \int E_p (a_p^{\rho_1*} a_p^{\rho_1} + b_p^{\rho_1*} b_p^{\rho_1}) d\mathbf{p}$$

$$\begin{aligned}
& + \frac{1}{(2\pi)^6} \sum_{i, \rho} \frac{g}{M^2} \int d\mathbf{r} d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 \\
& \times [a_{\mathbf{p}_1}^{\rho_1*} D_{\rho_1 \rho_2}^i a_{\mathbf{p}_2}^{\rho_2} e^{-i(\mathbf{p}_1 - \mathbf{p}_2)\mathbf{r}} - a_{\mathbf{p}_1}^{\rho_1*} B_{\rho_1 \rho_2}^i b_{\mathbf{p}_2}^{\rho_2*} e^{-i(\mathbf{p}_1 + \mathbf{p}_2)\mathbf{r}} \\
& + b_{\mathbf{p}_1}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}_2}^{\rho_2} e^{i(\mathbf{p}_1 + \mathbf{p}_2)\mathbf{r}} - b_{\mathbf{p}_1}^{\rho_1*} D_{\rho_1 \rho_2}^i b_{\mathbf{p}_2}^{\rho_2} e^{-i(\mathbf{p}_1 - \mathbf{p}_2)\mathbf{r}}] \\
& \times [a_{\mathbf{p}_3}^{\rho_3*} D_{\rho_3 \rho_4}^i a_{\mathbf{p}_4}^{\rho_4} e^{-i(\mathbf{p}_3 - \mathbf{p}_4)\mathbf{r}} - a_{\mathbf{p}_3}^{\rho_3*} B_{\rho_3 \rho_4}^i b_{\mathbf{p}_4}^{\rho_4*} e^{-i(\mathbf{p}_3 + \mathbf{p}_4)\mathbf{r}} \\
& + b_{\mathbf{p}_3}^{\rho_3} C_{\rho_3 \rho_4}^i a_{\mathbf{p}_4}^{\rho_4} e^{i(\mathbf{p}_3 + \mathbf{p}_4)\mathbf{r}} - b_{\mathbf{p}_3}^{\rho_3*} D_{\rho_3 \rho_4}^i b_{\mathbf{p}_4}^{\rho_4} e^{-i(\mathbf{p}_3 - \mathbf{p}_4)\mathbf{r}}], \quad (25)
\end{aligned}$$

where

$$\begin{aligned}
B_{p,q}^i &= (\tau_i \tau_q) \left( \sigma_2 - \frac{\boldsymbol{\sigma} \cdot \mathbf{p}}{E_p + M} \cdot \frac{\boldsymbol{\sigma} \cdot \mathbf{q}}{E_q + M} \sigma_2 \right) D_p D_q \\
C_{p,q}^i &= (\tau_i \tau_q) \left( \sigma_2 - \sigma_2 \frac{\boldsymbol{\sigma} \cdot \mathbf{p}}{E_p + M} \cdot \frac{\boldsymbol{\sigma} \cdot \mathbf{q}}{E_q + M} \right) D_p D_q \\
D_{p,q}^i &= \tau_i \left( \frac{\boldsymbol{\sigma} \cdot \mathbf{p}}{E_p + M} - \frac{\boldsymbol{\sigma} \cdot \mathbf{q}}{E_q + M} \right) D_p D_q \\
D_p &= \sqrt{(E_p + M)/2E_p},
\end{aligned}$$

and the subsidiary condition

$$A_k^i \Psi_0 = 0, \quad (21')$$

we perform the following canonical transformation

$$e^{iS} O e^{-iS} = O + [iS, O] + \frac{1}{2} [iS, [iS, O]] + \dots, \quad (26)$$

where

$$\begin{aligned}
S &= -\frac{f}{(2\pi)^{3/2}} \sum_j \int d\mathbf{k} d\mathbf{p} \frac{1}{\sqrt{2\omega_k}} \left[ \left( \frac{a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}-\mathbf{k}/2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} + \frac{b_{\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \right) A_{\mathbf{k}}^j \right. \\
& \quad \left. + \left( \frac{a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}+\mathbf{k}/2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} + \frac{b_{\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}+\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right) A_{\mathbf{k}}^j \right]. \quad (27)
\end{aligned}$$

We first consider the transformed form of (21'). Making the same approximation that leads to commutation relations (5) and (6), we obtain

$$\begin{aligned}
& \left[ A_{\mathbf{k}}^i + \frac{-if}{(2\pi)^{3/2}} \int d\mathbf{p} \frac{1}{\sqrt{2\omega_k}} \left( \frac{a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^i b_{\mathbf{p}+\mathbf{k}/2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} + \frac{b_{\mathbf{p}+\mathbf{k}/2}^{\rho_1} C_{\rho_1 \rho_2}^i a_{\mathbf{p}+\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right) \right. \\
& \quad \left. + \left\{ \frac{f^2}{(2\pi)^3} \frac{1}{\omega_k} \int d\mathbf{p} \left( \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k)^2} - \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k)^2} \right) \right\} A_{\mathbf{k}}^i \right] \Psi_0' = 0. \quad (28)
\end{aligned}$$

In (28), we adjust the value of  $f$  so that the term containing  $A_k^i$  is eliminated, i. e.

$$1 + \frac{f^2}{(2\pi)^3} \frac{1}{\omega_k} \int d\mathbf{p} \left( \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k)^2} - \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} - E_{\mathbf{p}-\mathbf{k}/2} - \omega_k)^2} \right) = 0.$$

From this equation  $f$  is fixed as  $f \sim 14.6$  with the use of cut-off as before. Then (28) becomes

$$\int d\mathbf{p} \frac{1}{\sqrt{2\omega_k}} \left( \frac{a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1*} B_{\rho_1\rho_2}^i b_{(\mathbf{p}+\mathbf{k}/2)}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} + \frac{b_{(\mathbf{p}-\mathbf{k}/2)}^{\rho_1} C_{\rho_1\rho_2}^i a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} - E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right) q_s^i = 0. \quad (29)$$

Next we transform the total Hamiltonian by (26). Before we do this, we rewrite  $H'$  as follows,

$$H' \sim \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 \{ [\text{I}] + [\text{II}] + [\text{III}] \\ + 2([\text{IV}] + [\text{V}] + [\text{VI}] + [\text{VII}]) \},$$

where

$$\begin{aligned} [\text{I}] &= a_{\mathbf{p}_3}^{\rho_3*} a_{\mathbf{p}_1}^{\rho_1*} a_{\mathbf{p}_2}^{\rho_2} a_{\mathbf{p}_4}^{\rho_4} D_{\rho_1\rho_2}^i D_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 - \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) \\ [\text{II}] &= a_{\mathbf{p}_3}^{\rho_3*} a_{\mathbf{p}_1}^{\rho_1*} b_{\mathbf{p}_2}^{\rho_2*} b_{\mathbf{p}_4}^{\rho_4*} B_{\rho_1\rho_2}^i B_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) \\ [\text{III}] &= b_{\mathbf{p}_3}^{\rho_3} b_{\mathbf{p}_1}^{\rho_1} a_{\mathbf{p}_2}^{\rho_2} a_{\mathbf{p}_4}^{\rho_4} C_{\rho_1\rho_2}^i C_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) \\ [\text{IV}] &= -a_{\mathbf{p}_1}^{\rho_1*} a_{\mathbf{p}_3}^{\rho_3*} b_{\mathbf{p}_4}^{\rho_4*} a_{\mathbf{p}_2}^{\rho_2} D_{\rho_1\rho_2}^i B_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 - \mathbf{p}_2 + \mathbf{p}_3 + \mathbf{p}_4) \\ [\text{V}] &= a_{\mathbf{p}_1}^{\rho_1*} b_{\mathbf{p}_3}^{\rho_3} a_{\mathbf{p}_4}^{\rho_4} a_{\mathbf{p}_2}^{\rho_2} D_{\rho_1\rho_2}^i C_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 - \mathbf{p}_2 - \mathbf{p}_3 - \mathbf{p}_4) \\ [\text{VI}] &= -a_{\mathbf{p}_1}^{\rho_1*} b_{\mathbf{p}_3}^{\rho_3*} b_{\mathbf{p}_4}^{\rho_4} a_{\mathbf{p}_2}^{\rho_2} D_{\rho_1\rho_2}^i D_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 - \mathbf{p}_2 + \mathbf{p}_3 - \mathbf{p}_4) \\ [\text{VII}] &= -a_{\mathbf{p}_1}^{\rho_1*} b_{\mathbf{p}_2}^{\rho_2*} b_{\mathbf{p}_3}^{\rho_3} a_{\mathbf{p}_4}^{\rho_4} B_{\rho_1\rho_2}^i C_{\rho_3\rho_4}^i \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{p}_3 - \mathbf{p}_4). \end{aligned} \quad (30)$$

Here we write only terms relevant to the following calculations. We transform (30) by (26) and pick up terms which correspond to the free meson Hamiltonian (we denote this by  $H_M$ ),

$$\begin{aligned} & \frac{1}{2} [i\mathcal{S}, [i\mathcal{S}, H_0]] \\ & \sim \frac{f^2}{(2\pi)^3} \sum_j \int dk d\mathbf{p} \frac{1}{2\omega_k} \cdot 4 (E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2}) \left\{ \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k)^2} \right. \\ & \quad \left. + \frac{1}{(E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k)^2} \right\} A_{\mathbf{k}}^{j*} A_{\mathbf{k}}^j, \end{aligned}$$

$$\begin{aligned}
& \frac{1}{2} [iS, [iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 ([\text{II}] + [\text{III}])] ] \\
& \rightarrow -2 \frac{f^2 g}{(2\pi)^6} \sum_j \int d\mathbf{k} d\mathbf{p} d\mathbf{p}' \frac{1}{2\omega_k} \frac{4+16}{M^2} \\
& \quad \times \left\{ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}/2} + E_{\mathbf{p}'-\mathbf{k}/2} + \omega_k} \right. \\
& \quad \left. + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}/2} + E_{\mathbf{p}'-\mathbf{k}/2} - \omega_k} \right\} A_{\mathbf{k}}^{j*} A_{\mathbf{k}}^j, \\
& \frac{1}{2} [iS, [iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 \cdot 2[\text{VII}]]] \\
& \rightarrow -2 \frac{f^2 g}{(2\pi)^6} \sum_j \int d\mathbf{k} d\mathbf{p} d\mathbf{p}' \frac{1}{2\omega_k} \frac{16}{M^2} \\
& \quad \times \left\{ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}/2} + E_{\mathbf{p}'-\mathbf{k}/2} - \omega_k} \right. \\
& \quad \left. + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}/2} + E_{\mathbf{p}'-\mathbf{k}/2} + \omega_k} \right\} A_{\mathbf{k}}^{j*} A_{\mathbf{k}}^j.
\end{aligned}$$

Making the approximation  $E_p \sim M$  and using the values,  $f=14.6$  and  $g=6.6$  and cutting off integrals with respect to  $\mathbf{p}$  and  $\mathbf{p}'$  at the nucleon mass,  $H_M$  becomes

$$H_M \sim \sum_j \int d\mathbf{k} \omega_k A_{\mathbf{k}}^{j*} A_{\mathbf{k}}^j. \quad (31)$$

Then we pick up meson-nucleon interaction terms corresponding to the  $p$ -wave interaction and  $s$ -wave interaction separately. First the  $p$ -wave interaction terms are

$$\begin{aligned}
& [iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 2([\text{IV}] + [\text{V}])] \\
& \cong \frac{-ifg}{(2\pi)^{3/2}} \frac{2}{M^2} \frac{1}{(2\pi)^3} \sum_j \int d\mathbf{k} d\mathbf{p} \frac{1}{\sqrt{2\omega_k}} \\
& \quad \times \left[ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \left\{ a_{\mathbf{p}_3}^* \tau_j \left( \frac{\boldsymbol{\sigma} \cdot (\mathbf{p}-\mathbf{k}/2)}{E_{\mathbf{p}-\mathbf{k}/2} + M} - \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_2}{E_{\mathbf{p}_2} + M} \right) a_{\mathbf{p}_2} d\mathbf{p}_3 d\mathbf{p}_2 \delta(\mathbf{p}_3 - \mathbf{p}_2 - \mathbf{k}) \right. \right. \\
& \quad \left. \left. + 4a_{\mathbf{p}_1}^* \tau_j \left( \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_1}{E_{\mathbf{p}_1} + M} - \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_2}{E_{\mathbf{p}_2} + M} \right) a_{\mathbf{p}_2} d\mathbf{p}_1 d\mathbf{p}_2 \delta(\mathbf{p}_1 - \mathbf{p}_2 - \mathbf{k}) \right\} A_{\mathbf{k}}^j \right. \\
& \quad \left. + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \left\{ a_{\mathbf{p}_3}^* \tau_j \left( \frac{\boldsymbol{\sigma} \cdot (\mathbf{p}+\mathbf{k}/2)}{E_{\mathbf{p}+\mathbf{k}/2} + M} - \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_2}{E_{\mathbf{p}_2} + M} \right) a_{\mathbf{p}_2} d\mathbf{p}_3 d\mathbf{p}_2 \delta(\mathbf{p}_3 - \mathbf{p}_2 + \mathbf{k}) \right. \right. \\
& \quad \left. \left. + 4a_{\mathbf{p}_1}^* \tau_j \left( \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_1}{E_{\mathbf{p}_1} + M} - \frac{\boldsymbol{\sigma} \cdot \mathbf{p}_2}{E_{\mathbf{p}_2} + M} \right) a_{\mathbf{p}_2} d\mathbf{p}_1 d\mathbf{p}_2 \delta(\mathbf{p}_1 - \mathbf{p}_2 + \mathbf{k}) \right\} A_{\mathbf{k}}^{j*} \right]
\end{aligned}$$



$$\begin{aligned}
& + \frac{1}{E_{p+k/2} + E_{p-k/2} - \omega_k} \left\{ a_{p_1}^* \tau_j \left( \frac{\sigma \cdot p_1}{E_{p_1} + M} - \frac{\sigma \cdot (p+k/2)}{E_{p+k/2} + M} \right) a_{p_4} d p_1 d p_4 \delta(p_1 - p_4 - k) \right. \\
& \quad \left. + 4 a_{p_1}^* \tau_j \left( \frac{\sigma \cdot p_1}{E_{p_1} + M} - \frac{\sigma \cdot p_2}{E_{p_2} + M} \right) a_{p_2} d p_1 d p_2 \delta(p_1 - p_2 - k) \right\} A_k^j \\
& + \frac{1}{E_{p+k/2} + E_{p-k/2} + \omega_k} \left\{ a_{p_1}^* \tau_j \left( \frac{\sigma \cdot p_1}{E_{p_1} + M} - \frac{\sigma \cdot (p-k/2)}{E_{p-k/2} + M} \right) a_{p_4} d p_1 d p_4 \delta(p_1 - p_4 + k) \right. \\
& \quad \left. + 4 a_{p_1}^* \tau_j \left( \frac{\sigma \cdot p_1}{E_{p_1} + M} - \frac{\sigma \cdot p_2}{E_{p_2} + M} \right) a_{p_2} d p_1 d p_2 \delta(p_1 - p_2 + k) \right\} A_k^j \Big].
\end{aligned}$$

Making the approximation  $E_p \sim M$ , the above expression becomes

$$\begin{aligned}
H_{\gamma\pi} & \sim \frac{iG}{\mu} \frac{1}{(2\pi)^{3/2}} \sum_j \int d p_1 d p_2 d k \frac{1}{\sqrt{2\omega_k}} \\
& \times a_{p_1}^* \tau_j (\sigma \cdot k) \left\{ -A_k^j \delta(p_1 - p_2 - k) + A_k^{j*} \delta(p_1 - p_2 + k) \right\} a_{p_2}, \quad (32)
\end{aligned}$$

where

$$\frac{G^2}{4\pi} = \frac{(2 \cdot 3 \pi^2 \cdot \mu \cdot M \cdot f(t))^2}{4\pi} \sim 0.08.$$

Here we denote the  $p$ -wave interaction Hamiltonian by  $H_{\gamma\pi}$  and use the values,  $f=14.6$  and  $g=6.6$ , and the cut-off procedure as before.

Secondly, we pick up  $s$ -wave interaction terms,

$$\begin{aligned}
& [iS, H_0] \\
& = \frac{if}{(2\pi)^{3/2}} \sum_j \int d k d p \frac{1}{\sqrt{2\omega_k}} (E_{p+k/2} + E_{p-k/2}) \\
& \times \left[ \left\{ -\frac{a_{p+k/2}^{\rho_1*} B_{\rho_1\rho_2}^j b_{\rho_2}^{\rho_2*}(p-k/2)}{E_{p+k/2} + E_{p-k/2} - \omega_k} + \frac{b_{\rho_1}^{\rho_1}(p+k/2) C_{\rho_1\rho_2}^j a_{p-k/2}^{\rho_2}}{E_{p+k/2} + E_{p-k/2} + \omega_k} \right\} A_k^j \right. \\
& \quad \left. + \left\{ -\frac{a_{p-k/2}^{\rho_1*} B_{\rho_1\rho_2}^j b_{\rho_2}^{\rho_2*}(p+k/2)}{E_{p+k/2} + E_{p-k/2} + \omega_k} + \frac{b_{\rho_1}^{\rho_1}(p-k/2) C_{\rho_1\rho_2}^j a_{p+k/2}^{\rho_2}}{E_{p+k/2} + E_{p-k/2} - \omega_k} \right\} A_k^j \right], \quad (33a)
\end{aligned}$$

$$\begin{aligned}
& \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d p_1 d p_2 d p_3 d p_4 [\Pi] \right] \\
& \sim \frac{ifg}{(2\pi)^{3/2}} \sum_j \frac{2}{M^2} \int d p_1 d p_2 d k d p \frac{5}{\sqrt{2\omega_k}} \\
& \times \left\{ \frac{a_{p+k/2}^{\rho_1*} B_{\rho_1\rho_2}^j b_{\rho_2}^{\rho_2*}}{E_{p+k/2} + E_{p-k/2} + \omega_k} A_k^j \delta(p_1 + p_2 - k) + \frac{a_{p+k/2}^{\rho_1*} B_{\rho_1\rho_2}^j b_{\rho_2}^{\rho_2*}}{E_{p+k/2} + E_{p-k/2} - \omega_k} \right. \\
& \quad \left. \times A_k^{j*} \delta(p_1 + p_2 + k) \right\}, \quad (33b)
\end{aligned}$$

$$\begin{aligned}
& \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 [\text{III}] \right] \\
& \rightarrow \frac{-ifg}{(2\pi)^{9/2}} \sum_j \frac{2}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{k} d\mathbf{p} \frac{5}{\sqrt{2\omega_k}} \\
& \quad \times \left\{ \frac{b_{\mathbf{p}_1}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}_2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} A_{\mathbf{k}}^j \delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{k}) + \frac{b_{\mathbf{p}_1}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}_2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \right. \\
& \quad \left. \times A_{\mathbf{k}}^{j*} \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{k}) \right\}, \quad (33c)
\end{aligned}$$

$$\begin{aligned}
& \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 \cdot 2 [\text{VII}] \right] \\
& \rightarrow \frac{ifg}{(2\pi)^{9/2}} \sum_j \frac{2}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{k} d\mathbf{p} \frac{4}{\sqrt{2\omega_k}} \\
& \quad \times \left[ \left\{ \frac{a_{\mathbf{p}_1}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}_2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{k}) - \frac{b_{\mathbf{p}_1}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}_2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \right. \right. \\
& \quad \left. \left. \times \delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{k}) \right\} A_{\mathbf{k}}^j \right. \\
& \quad \left. + \left\{ \frac{a_{\mathbf{p}_1}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}_2}^{\rho_2*}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \delta(\mathbf{p}_1 + \mathbf{p}_2 + \mathbf{k}) - \frac{b_{\mathbf{p}_1}^{\rho_1} C_{\rho_1 \rho_2}^j a_{\mathbf{p}_2}^{\rho_2}}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right. \right. \\
& \quad \left. \left. \times \delta(\mathbf{p}_1 + \mathbf{p}_2 - \mathbf{k}) \right\} A_{\mathbf{k}}^{j*} \right], \quad (33d)
\end{aligned}$$

$$\begin{aligned}
& \frac{1}{2} [iS, [iS, H_0]] \\
& \rightarrow \frac{-f^2}{(2\pi)^3} \sum_{ij} \int d\mathbf{k} d\mathbf{p} d\mathbf{k}' d\mathbf{p}' \frac{1}{\sqrt{2\omega_k}} \frac{1}{\sqrt{2\omega_{k'}}} (E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2}) \\
& \quad \times \left\{ \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} - \omega_{k'}} \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_k} \right. \\
& \quad \times a_{\mathbf{p}+\mathbf{k}/2}^* \tau_j \tau_i a_{\mathbf{p}'+\mathbf{k}'/2} A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j \delta\left(\mathbf{p} - \mathbf{p}' - \frac{\mathbf{k}}{2} + \frac{\mathbf{k}'}{2}\right) \\
& \quad + \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} + \omega_{k'}} \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_k} \\
& \quad \left. \times a_{\mathbf{p}'-\mathbf{k}'/2}^* \tau_j \tau_i a_{\mathbf{p}+\mathbf{k}/2} A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j \delta\left(\mathbf{p} - \mathbf{p}' + \frac{\mathbf{k}}{2} - \frac{\mathbf{k}'}{2}\right) \right\}, \quad (34a)
\end{aligned}$$

$$\frac{1}{2} \left[ iS, \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 [\text{II}] \right] \right]$$

$$\begin{aligned}
& \rightarrow \frac{f^2}{(2\pi)^3} \sum_{ij} \frac{2}{M^2} \int dk d\mathbf{p}_1 dk' d\mathbf{p}' d\mathbf{q} \frac{g}{(2\pi)^3} \frac{1}{\sqrt{2\omega_k}} \frac{1}{\sqrt{2\omega_{k'}}} 5 A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j \\
& \times \left[ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} - \omega_{\mathbf{k}'}} \right. \\
& \times \left\{ a_{\mathbf{p}_1}^* \tau_j \tau_i a_{\mathbf{p}'+\mathbf{k}'/2} \delta\left(\mathbf{p}_1 - \mathbf{p}' - \mathbf{k} + \frac{\mathbf{k}'}{2}\right) \right. \\
& \left. + a_{\mathbf{p}_1}^* \tau_i \tau_j a_{\mathbf{p}-\mathbf{k}/2} \delta\left(\mathbf{p}_1 - \mathbf{p} - \frac{\mathbf{k}}{2} + \mathbf{k}'\right) \right\} \\
& + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} + \omega_{\mathbf{k}'}} \\
& \times \left\{ a_{\mathbf{p}_1}^* \tau_j \tau_i a_{\mathbf{p}'+\mathbf{k}'/2} \delta\left(\mathbf{p}_1 - \mathbf{p}' - \mathbf{k} + \frac{\mathbf{k}'}{2}\right) \right. \\
& \left. + a_{\mathbf{p}_1}^* \tau_i \tau_j a_{\mathbf{p}-\mathbf{k}/2} \delta\left(\mathbf{p}_1 - \mathbf{p} - \frac{\mathbf{k}}{2} + \mathbf{k}'\right) \right\} \Bigg], \tag{34b}
\end{aligned}$$

$$\begin{aligned}
& \frac{1}{2} \left[ iS, \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 [\text{III}] \right] \right] \\
& \rightarrow \frac{f^2}{(2\pi)^3} \sum_{ij} \frac{2}{M^2} \int dk d\mathbf{p}_4 dk' d\mathbf{p}' d\mathbf{q} \frac{g}{(2\pi)^3} \frac{1}{\sqrt{2\omega_k}} \frac{1}{\sqrt{2\omega_{k'}}} 5 A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j \\
& \times \left[ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} - \omega_{\mathbf{k}'}} \right. \\
& \times \left\{ a_{\mathbf{p}'-\mathbf{k}'/2}^* \tau_i \tau_j a_{\mathbf{p}_4} \delta\left(\mathbf{p}_4 - \mathbf{p}' - \frac{\mathbf{k}'}{2} + \mathbf{k}\right) \right. \\
& \left. + a_{\mathbf{p}+\mathbf{k}/2}^* \tau_j \tau_i a_{\mathbf{p}_4} \delta\left(\mathbf{p}_4 - \mathbf{p} - \mathbf{k}' + \frac{\mathbf{k}}{2}\right) \right\} \\
& + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} + \omega_{\mathbf{k}'}} \\
& \times \left\{ a_{\mathbf{p}'-\mathbf{k}'/2}^* \tau_i \tau_j a_{\mathbf{p}_4} \delta\left(\mathbf{p}_4 - \mathbf{p}' - \frac{\mathbf{k}'}{2} + \mathbf{k}\right) \right. \\
& \left. + a_{\mathbf{p}+\mathbf{k}/2}^* \tau_j \tau_i a_{\mathbf{p}_4} \delta\left(\mathbf{p}_4 - \mathbf{p} - \mathbf{k}' + \frac{\mathbf{k}}{2}\right) \right\} \Bigg], \tag{34c}
\end{aligned}$$

$$\begin{aligned}
& \frac{1}{2} \left[ iS, \left[ iS, \frac{1}{(2\pi)^3} \sum_{i\rho} \frac{g}{M^2} \int d\mathbf{p}_1 d\mathbf{p}_2 d\mathbf{p}_3 d\mathbf{p}_4 2[\text{VII}] \right] \right] \\
& \rightarrow \frac{f^2}{(2\pi)^3} \sum_{ij} \frac{2}{M^2} \int dk d\mathbf{p}' dk' d\mathbf{p}_4 d\mathbf{q} \frac{g}{(2\pi)^3} \frac{1}{\sqrt{2\omega_k}} \frac{1}{\sqrt{2\omega_{k'}}} A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j
\end{aligned}$$

$$\begin{aligned}
& \times \left[ \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} + \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} + \omega_{\mathbf{k}'}} \right. \\
& \times \left\{ 8a_{\mathbf{p}'-\mathbf{k}'/2}^* \tau_i \tau_j a_{\mathbf{p}_4} \delta \left( \mathbf{p}_4 - \mathbf{p}' - \frac{\mathbf{k}'}{2} + \mathbf{k} \right) \right. \\
& \left. - a_{\mathbf{p}_1}^* (4 - \tau_i \tau_j) a_{\mathbf{p}_4} \delta (\mathbf{p}_1 - \mathbf{p}_4 - \mathbf{k} - \mathbf{k}') \right\} \\
& + \frac{1}{E_{\mathbf{p}+\mathbf{k}/2} + E_{\mathbf{p}-\mathbf{k}/2} - \omega_{\mathbf{k}}} \cdot \frac{1}{E_{\mathbf{p}'+\mathbf{k}'/2} + E_{\mathbf{p}'-\mathbf{k}'/2} - \omega_{\mathbf{k}'}} \\
& \times \left\{ 8a_{\mathbf{p}_1}^* \tau_j \tau_i a_{\mathbf{p}'+\mathbf{k}'/2} \delta \left( \mathbf{p}_1 - \mathbf{p}' + \frac{\mathbf{k}'}{2} - \mathbf{k} \right) \right. \\
& \left. - a_{\mathbf{p}_1}^* (4 - \tau_i \tau_j) a_{\mathbf{p}_4} \delta (\mathbf{p}_1 - \mathbf{p}_4 + \mathbf{k}' - \mathbf{k}) \right\} \Big]. \quad (34d)
\end{aligned}$$

Making the approximation,  $E_p \sim M$  and using the value  $g=6.6$  and the cut-off procedure, (33a), (33b), (33c) and (33d) become

$$\begin{aligned}
& \frac{if}{(2\pi)^{3/2}} \sum_j \int d\mathbf{k} d\mathbf{p} \frac{1}{V \sqrt{2\omega_{\mathbf{k}}}} \left[ x (a_{\mathbf{p}+\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}-\mathbf{k}/2}^{\rho_2*} A_{\mathbf{k}-\mathbf{b}^{\rho_1}(\mathbf{p}-\mathbf{k}/2)}^j - b_{\mathbf{p}-\mathbf{k}/2}^{\rho_2*} C_{\rho_1 \rho_2}^j a_{\mathbf{p}+\mathbf{k}/2}^{\rho_2} A_{\mathbf{k}}^{i*}) \right. \\
& \left. - y (a_{\mathbf{p}-\mathbf{k}/2}^{\rho_1*} B_{\rho_1 \rho_2}^j b_{\mathbf{p}+\mathbf{k}/2}^{\rho_2*} A_{\mathbf{k}}^{i*} - b_{\mathbf{p}+\mathbf{k}/2}^{\rho_2*} C_{\rho_1 \rho_2}^j a_{\mathbf{p}-\mathbf{k}/2}^{\rho_2} A_{\mathbf{k}}^j) \right],
\end{aligned}$$

$$x \sim y \sim -0.05$$

and this is negligible compared with the others. This fact shows that the above transformation separate the  $\pi$ -meson mode in good approximation. (34a), (34b), (34c) and (34d) become in the same approximation

$$H_s \sim \frac{f^2}{(2\pi)^3} \sum_{ij} \int d\mathbf{p} d\mathbf{p}' d\mathbf{k} d\mathbf{k}' \frac{1}{V \sqrt{2\omega_{\mathbf{k}}}} \frac{1}{V \sqrt{2\omega_{\mathbf{k}'}}} \frac{1}{2M} a_{\mathbf{p}'}^* a_{\mathbf{p}} A_{\mathbf{k}'}^{i*} A_{\mathbf{k}}^j \delta(\mathbf{p}' + \mathbf{k}' - \mathbf{p} - \mathbf{k}). \quad (35)$$

Here we denote the effective  $s$ -wave interaction by  $H_s$ .

In this way, if we choose suitable parameters, we can obtain the free meson Hamiltonian, effective  $p$ -wave interaction and  $s$ -wave core interaction. (31), (32) and (35) show that the effective pion-nucleon interaction obtained from our interaction Hamiltonian (1) is equivalent to the customary pseudoscalar meson theory with pseudoscalar coupling. Although we obtain our results on the basis of one pair approximation, the above conclusion will hold even if we take account of higher configurations, because in the new Tamm-Dancoff method the lowest approximation includes at least partially the effects of infinite number of virtual nucleon-antinucleon pairs and the cut-off procedure makes contributions from terms neglected here rather small.



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# Vibrational Thermodynamic Properties of Lattices with Defects, I

—The Linear Lattice\*—

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In this paper, several methods of analysing vibrational properties of crystal lattices with defects are developed. Integral expressions for additive functions of normal mode frequencies are derived following the work of Montroll and his collaborators. It is shown that the Helmholtz free energy can be evaluated at high and low temperatures without performing the integrations. The methods presented are valid for lattices of all odd dimensions, although specific results are presented here for one-dimensional monatomic and diatomic lattices. Using a method similar to that developed by Lifshitz, it is shown that the properties of a lattice with defects can be expanded in a series of powers of the concentration of defects. The coefficient of the  $n^{\text{th}}$  power depends on the properties of a lattice with  $n$  defects. Examples of such expansions are given. An exact expression for the frequency distribution function of a monatomic linear chain with an isotope defect is given.

## § 1. Introduction

In the study of the effect of defects on the thermodynamic properties of crystals and order-disorder phenomena it has been customary to ignore the vibrational contribution to the free energy and to study static effects only. This reduces the problem to a purely configurational one. Many methods of solution for these configurational problems have been devised. However, it is known both experimentally and theoretically that the vibrational contribution of defects and disorder to thermodynamic functions of crystals is not negligible. We therefore present in this paper a number of methods which have been found useful in studying defect problems. We illustrate the use of these techniques by calculating several quantities of interest for the one-dimensional lattice. Although there is very little physical significance to such quantities as the specific heat or free energy of a one-dimensional lattice, the mathematical tools that we use in the following calculations can be applied to three-dimensional problems as well. The one-dimensional calculations

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have the virtue of mathematical simplicity yet serve to illustrate these methods whose exposition is the primary purpose of the present paper. Calculations for the more realistic three-dimensional cases will be described in future papers.

The earliest calculation of the effect of large numbers of defects on the vibrational properties of crystals is due to Stripp and Kirkwood<sup>1</sup>. They determined the effect of lattice vacancies on the vibrational partition function to second order in perturbation theory. They showed that the vibrational contribution effectively lowers the energy for vacancy formation so that the equilibrium vacancy concentration should exceed that of a static model. They also showed that this effect is more pronounced at higher temperatures. Furthermore the inclusion of the vibrational contribution approximately halves the "melting temperature" calculated according to the hole theory. It is difficult to estimate the quantitative accuracy of their results since the use of perturbation theory neglects the contributions from any impurity frequencies associated with the defects. These may be of the same order of magnitude as arise from the in-band frequency shifts. The present work is an extension of methods first introduced in defect calculations by Montroll and his collaborators<sup>2,3,4,5</sup>, and takes into account both in-band frequency shifts and impurity frequencies.

## § 2. Mathematical preliminaries

In this paper we will be concerned with the evaluation of various additive functions of the normal mode frequencies of a one-dimensional lattice. These functions can be expressed as

$$S = \sum_j f(\omega_j) \quad (2.1)$$

where the normal mode frequencies  $\omega_j$  are the roots of a determinantal equation which we write as

$$|M(\omega)| = 0. \quad (2.2)$$

The zero-point energy of the lattice is given by  $S$  when  $f(z) = \frac{1}{2}\hbar z$ ; the Fourier transform of the frequency spectrum corresponds to  $f(z) = N^{-1} \exp(i a z)$ , where  $N$  is the total number of degrees of freedom in the lattice; and the Helmholtz free energy corresponds to  $f(z) = kT \ln \{2 \sinh (\hbar z / 2kT)\}$ .

Using a well-known result from the theory of contour integration we can express  $S$  in an alternative way as

$$S = -\frac{1}{2\pi i} \int_C f(z) d \ln |M(z)| \quad (2.4)$$

where  $C$  is any closed, counterclockwise contour which encloses all the zeroes of  $M(z)$  but none of the poles of  $f(z)$ .

When the lattice contains defects we can write  $M(\omega)$ , the matrix of the secular determinant, as the sum of two square matrices

$$M(\omega) = M_0(\omega) + \delta M(\omega) \quad (2.5)$$

where  $M_0(\omega)$  is the matrix of the secular determinant for the lattice containing no defects, and  $\delta M(\omega)$  represents the perturbation due to the addition of the defects. We can now express  $M(\omega)$  in a form more convenient for our purposes by writing

$$\begin{aligned} M(\omega) &= M_0(\omega) [I + M_0^{-1}(\omega) \delta M(\omega)] \\ &= M_0(\omega) [I + D(\omega)] = M_0(\omega) J(\omega). \end{aligned} \quad (2.6)$$

These equations define the matrices  $D(\omega)$  and  $J(\omega)$ . Since the determinant of a product of two matrices is equal to the product of the determinants of the individual matrices, we have

$$|M(\omega)| = |M_0(\omega)| |J(\omega)|. \quad (2.7)$$

Thus the impurity frequencies are roots of the equation  $|J(\omega)| = 0$ . Substituting the factorization of  $M(\omega)$  into Eq. (2.4) we obtain finally the following expression for  $\Delta S$ , the change in  $S$  due to the presence of defects in the lattice

$$\Delta S = \frac{1}{2\pi i} \int_c f(z) d \ln |J(z)|. \quad (2.8)$$

The change in an additive function  $S$  which results from a single defect at some point  $\alpha$  is

$$\Delta S_\alpha = \frac{1}{2\pi i} \int_c f(z) d \ln |J_\alpha(z)| \quad (2.9)$$

and might be referred to as the "self- $S$ " of the defect. The "interaction- $S$ " of a pair of defects located at  $\alpha$  and  $\beta$  respectively is defined as the difference between the  $S$  of a system of two interacting defects and that of a pair of isolated defects and is given by

$$\Delta S_{\alpha,\beta} = \frac{1}{2\pi i} \int_c f(z) d \ln \frac{|J_{\alpha,\beta}(z)|}{|J_\alpha(z)| |J_\beta(z)|}. \quad (2.10)$$

It can be seen from eq. (2.8) that the matrix  $J(\omega)$  is of central importance in the calculation of any of the quantities  $\Delta S$ . We shall also show that  $J(\omega)$  can be used as a moment generating function and also to find the low temperature behavior of the free energy. Thus we can often circumvent the integration implied by eq. (2.8) in finding high and low temperature expansions of thermodynamic properties which depend on the normal mode frequencies.

The formulae eqs. (2.8) – (2.10) are independent of the type of lattice being analyzed and its dimension. However, the application of these techniques to defect calculations requires a sufficiently simple model of a crystal lattice so that the elements of the inverse matrix [cf. eq. (2.6)]  $M_0^{-1}(\omega)$  are readily obtained. For this model we will take a simple cubic lattice with nearest neighbor interactions, both central and non-central. The difference equations for the amplitudes  $u_{n_1 n_2 n_3}$  are given by

$$M\omega^2 u_{n_1 n_2 n_3} + \gamma_1 (u_{n_1+1, n_2 n_3} - 2u_{n_1 n_2 n_3} + u_{n_1-1, n_2 n_3})$$



$$\begin{aligned}
 & +\gamma_2(u_{n_1 n_2+1, n_3}-2u_{n_1 n_2 n_3}+u_{n_1 n_2-1, n_3}) \\
 & +\gamma_3(u_{n_1 n_2 n_3+1}-2u_{n_1 n_2 n_3}+u_{n_1 n_2 n_3-1})=0,
 \end{aligned}
 \quad (2.11)$$

where  $M$  is the particle mass and the  $\gamma$ 's are the force constants. Under the assumption of cyclic boundary conditions the elements of the inverse matrix  $M^{-1}(\omega)$  are found to be<sup>3</sup>

$$\alpha_{ij}^{(-1)} = \frac{1}{N^3} \sum_{s_1} \sum_{s_2} \sum_{s_3} \frac{\cos 2\pi s \cdot (\mathbf{i}-\mathbf{j})/N}{M\omega^2 - 2\sum_n \gamma_n (1 - \cos \pi s_n/N)}. \quad (2.12)$$

The form in one dimension is exactly the same except that there is only a single  $s$  and a single force constant. In the limit as  $N \rightarrow \infty$ , the sums appearing in eq. (2.12) can be written as a triple integral:

$$a_{ij}^{(-1)} = \frac{1}{\pi^3} \iiint \frac{\cos(i_1-j_1)\varphi_1 \cos(i_2-j_2)\varphi_2 \cos(i_3-j_3)\varphi_3 d\varphi_1 d\varphi_2 d\varphi_3}{M\omega^2 - 2\gamma_1(1 - \cos\varphi_1) - 2\gamma_2(1 - \cos\varphi_2) - 2\gamma_3(1 - \cos\varphi_3)} \quad (2.13)$$

where the integral converges for  $\omega^2 > \omega_L^2$  and  $\omega^2 < 0$ , where  $\omega_L$  is the largest normal mode frequency.

In one dimension it is possible to evaluate the integral which appears here with the result that

$$a_{ij}^{(-1)} = -\frac{1}{M\omega_L^2} \frac{(\sqrt{f^2+1}-|f|)^{2|t-j|}}{|f| \sqrt{1+f^2}} \quad \omega^2 < 0 \quad (2.14)$$

where  $\omega_L^2 = 4\gamma M^{-1}$  and  $f = \omega/\omega_L$ . The matrix elements  $a_{ij}^{(-1)}$  for an alternating diatomic chain in which the even lattice sites are occupied by particles of mass  $M_1$  while the odd sites are occupied by particles of mass  $M_2$  are<sup>5)</sup>

$$a_{ij}^{(-1)} = -\frac{\omega_1\omega_2}{2\gamma} \frac{1}{|\omega|\sqrt{\omega^2+\omega_L^2}} \frac{[(\omega_1^2+\omega^2)^{1/2}(\omega_2^2+\omega^2)^{1/2} - (\omega^4+\omega^2\omega_L^2)^{1/2}]^{|t-j|}}{(\omega_1\omega_2)^{|t-j|}} \quad \omega^2 < 0 \quad (2.15)$$

where  $\omega_j^2 = 2\gamma M_j^{-1}$  and  $\omega_L^2 = \omega_1^2 + \omega_2^2$ .

The contour  $C$  which appears in eq. (2.8) is usually taken to be the  $D$  shaped contour shown in Fig. 1. It can be shown in most cases that the contribution from the circular part of the contour vanishes in the limit  $R \rightarrow \infty$  (however, see the appendix). The resulting integrals are usually quite difficult to evaluate; hence we will outline some alternative methods for finding high and low temperature expansions for thermodynamic quantities.

We begin by rewriting the expression for the free energy in the form

$$\begin{aligned}
 A &= \kappa T \sum_j \ln(2 \sinh \frac{\hbar \omega_j}{2\kappa T}) = \frac{\hbar}{2} \sum_j \omega_j + \kappa T \sum_j \ln(1 - e^{-\frac{\hbar \omega_j}{\kappa T}}) \\
 &= E_0 - \kappa T \sum_{n=1}^{\infty} I_n/n
 \end{aligned}
 \quad (2.16)$$

\* In the rest of the paper,  $a_{ij}^{(-1)}$  will be denoted by  $g(m)$  where  $m=|i-j|$ . The  $g(m)$  are the Green's functions discussed by Montroll and Potts<sup>2,3</sup>.

where  $E_0$  is the zero-point energy of the lattice and  $I_n$  is given by

$$I_n = \sum_j e^{-n\hbar\omega_j/kT} \quad (2.17)$$

We may now write for the change in the quantities  $\Delta E_0$  and  $\Delta I_n$

$$\begin{aligned} \Delta E_0 &= -\frac{\hbar\omega_L}{2\pi} \int_0^\infty f d \ln |\Delta(if)| \\ &= -\frac{\hbar\omega_L}{2\pi} \int_0^\infty f \Omega(f) df, \\ \Delta I_n &= \frac{1}{\pi} \int_0^\infty \sin\left(\frac{n\hbar\omega_L}{kT} f\right) \Omega(f) df \\ &= \frac{1}{\pi} \int_0^\infty \sin(\alpha_n f) \Omega(f) df \end{aligned} \quad (2.18)$$

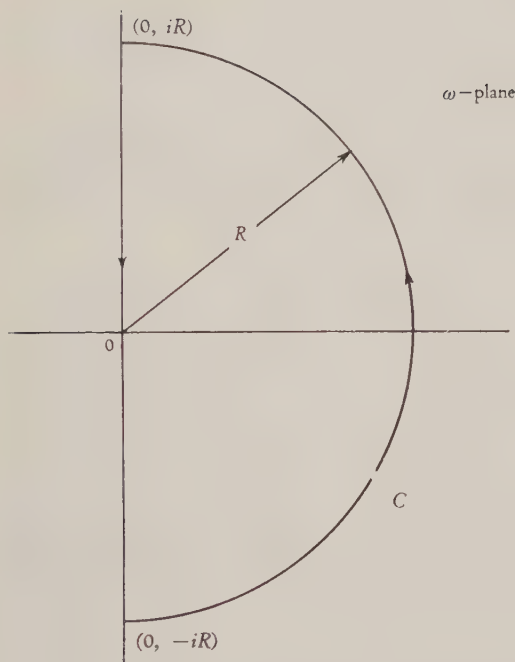


Figure 1. Contour of integration in the  $\omega$ -plane.

where we have used the notation  $\Omega(f) = (d/df) \ln |\Delta(if)|$  and  $\alpha_n = n\hbar\omega_L/kT$  and the fact that  $\Omega(f)$  is an odd function of  $f$ . At low temperatures  $\alpha_n$  will be large. Hence, if we integrate the expression for  $\Delta I_n$  by parts successively we find the following asymptotic expansion

$$\Delta I_n = \frac{1}{\pi} \left\{ \frac{\Omega(0)}{\alpha_n} - \frac{\Omega^{(2)}(0)}{\alpha_n^3} + \frac{\Omega^{(4)}(0)}{\alpha_n^5} - \dots \right\} \quad (2.19)$$

where we have assumed that  $\Omega(f)$  and all of its derivatives vanish in the limit  $f \rightarrow \infty$ . When we substitute this expansion into the expression for  $\Delta A$  we find that the change in the free energy is given by

$$\begin{aligned} \Delta A \sim \Delta E_0 - \frac{\kappa T}{\pi} \sum_{n=0}^{\infty} (-1)^n \frac{2^{2n+1} \pi^{2n+2}}{(2n+2)!} \\ \times \Omega^{(2n)}(0) \left( \frac{\kappa T}{\hbar\omega_L} \right)^{2n+1} B_{n+1} \end{aligned} \quad (2.20)$$

where  $B_n$  is the  $n$ 'th Bernoulli number. This expression is not limited to one dimension and shows that the low temperature change in free energy is related only to the behavior of  $\Omega(f)$  at the origin.

The high temperature expansion of the free energy is conveniently obtained by the method of moments. When the condition  $\frac{\hbar\omega}{\kappa T} < 2\pi$  we can expand the free energy as<sup>7)</sup>

$$A = \kappa T \sum_{j=1}^N \ln \frac{\hbar \omega_j}{\kappa T} - \kappa T \sum_{n=1}^{\infty} (-1)^n \frac{B_n \left( \frac{\hbar}{\kappa T} \right)^{2n}}{2n (2n)!} \sum_{j=1}^N \omega_j^{2n} \quad (2.21)$$

where, again, the  $B_n$  are Bernoulli numbers. The first term can be written in the form

$$\begin{aligned} \sum_{j=1}^N \ln \frac{\hbar \omega_j}{\kappa T} &= \ln \left( \frac{\hbar}{\kappa T} \right)^N + \frac{1}{2} \ln \prod_{j=1}^N \omega_j^2 \\ &= \ln \left( \frac{\hbar}{\kappa T} \right)^N + \frac{1}{2} \ln |D(0)| \end{aligned} \quad (2.22)$$

where  $|D(0)|$  is the determinant of the matrix  $D(0)$  whose eigenvalues are the  $\omega_j^2$ 's.\* We also have the relation

$$\sum_{j=1}^N \omega_j^{2n} = N \mu_{2n} = \text{Tr} [D(0)]^n \quad (2.23)$$

where  $\mu_{2n}$  is the  $2n$ 'th moment. We therefore obtain

$$JA = \frac{\kappa T}{2} \ln |D(0)| - \kappa T \sum_{n=1}^{\infty} \frac{(-1)^n B_n}{(2n) (2n)!} \left( \frac{\hbar}{\kappa T} \right)^{2n} N \mu_{2n} \quad (2.24)$$

where  $N \mu_{2n} = \text{Tr} [D(0)]^n - \text{Tr} [D_0(0)]^n$ . Hence in order to find a high temperature expansion of the free energy we need only find an expression for the moments. We shall show that this can be furnished if we know the function  $\Omega(f)$  introduced in eq. (2.18).

By definition  $\Omega(f)$  can be written in the form

$$\Omega(f) = \frac{d}{df} \sum_j \ln \left( \frac{\omega^2 + \nu_j^2}{\omega^2 + \nu_j^2(0)} \right) = 2\omega \omega_L \sum_j \frac{\nu_j^2(0) - \nu_j^2}{(\omega^2 + \nu_j^2)(\omega^2 + \nu_j^2(0))} \quad (2.25)$$

where the  $\nu_i(0)$  are the frequencies of the unperturbed lattice. If  $\omega$  is large enough we can expand this in a Taylor series in powers of  $f^{-1}$ ,

$$\Omega(f) = \frac{2}{f} \sum_{n=0}^{\infty} \sum_j \frac{(-1)^n}{\omega_L^{2n} f^{2n}} \{ \nu_j^{2n} - \nu_j^{2n}(0) \} = \frac{2N}{f} \sum_{n=1}^{\infty} \frac{(-1)^n \mu_{2n}}{\omega_L^{2n} f^{2n}} \quad (2.26)$$

Hence we see that  $\Omega(f)$  is really a moment difference generating function. If the coefficient of  $2(-1)^n f^{-(2n+1)}$  in the expansion of  $\Omega(f)$  is given by  $\alpha_{2n+1}$  then we can write for  $JA$ ,

$$JA = \frac{\kappa T}{2} \ln |D(0)| - \kappa T \sum_{n=1}^{\infty} \frac{B_n}{(2n) (2n)!} \left( \frac{\hbar \omega_L}{\kappa T} \right)^{2n} \alpha_{2n+1} \quad (2.27)$$

In the remainder of this paper we illustrate the methods presented in this section

\* The matrix  $D(0)$  is obtained from the matrix  $M(\omega)$  by dividing every row and column of  $M(\omega)$  by the square root of the mass appearing in the diagonal element corresponding to that row and column, and changing the sign of each term. The resulting matrix can be written as  $D(0) = \omega^2 I$  which defines  $D(0)$ . Note that  $D(0)$  is independent of  $\omega^2$ .

by calculating the changes in thermodynamic functions of one-dimensional lattices due to the presence of several kinds of defects.

### § 3. Effect of defects on the thermodynamic properties of a monatomic linear chain

In this section we consider the effect of the following kinds of defects on the vibrational free energy of a monatomic linear chain: (i) a defect which changes both the mass of a given particle as well as the force constants associated with the interactions joining it to its nearest neighbors; (ii) a single isotope defect; (iii) a pair of isotope defects; (iv) anomalous force constants; and (v) a lattice vacancy. All other thermodynamic functions can be obtained from the free energy by well known relations and hence will not be treated here.

#### (i) A General Impurity Atom

If a normal atom whose mass is  $M$  is replaced by an impurity atom whose mass is  $M'$ , and the force constants  $\gamma$  of its interaction with its nearest neighbors are altered to  $\gamma'$ , the defect determinant  $|A(\omega)|$  is found to be

$$\begin{aligned}
 |A(\omega)| = & \begin{vmatrix} 1 - \varepsilon_2 g(-1) - \varepsilon_1 g(0) & \varepsilon_1 [g(0) + g(-2)] + 2\varepsilon_2 g(-1) & -\varepsilon_2 g(-1) - \varepsilon_1 g(-2) \\ -\varepsilon_2 g(0) - \varepsilon_1 g(1) & 1 + \varepsilon_1 [g(1) + g(-1)] + 2\varepsilon_2 g(0) & -\varepsilon_2 g(0) - \varepsilon_1 g(-1) \\ -\varepsilon_2 g(1) - \varepsilon_1 g(2) & \varepsilon_1 [g(2) + g(0)] + 2\varepsilon_2 g(1) & 1 - \varepsilon_2 g(1) - \varepsilon_1 g(0) \end{vmatrix} \\
 & \quad (3.1)
 \end{aligned}$$

where  $\varepsilon_1 = \gamma' - \gamma$  and  $\varepsilon_2 = \gamma - \gamma' (M/M')$ . This determinant factors into

$$\begin{aligned}
 |A(\omega)| = & \{1 - (\gamma' - \gamma)[g(0) - g(2)]\} \{1 + (\gamma' - \gamma) \frac{M'}{\gamma'} \omega^2 g(1) \\
 & + \left( \frac{M'}{\gamma'} \gamma - M \right) \omega^2 g(0)\}. \quad (3.2)
 \end{aligned}$$

The change in the zero-point energy of the lattice is given by

$$\begin{aligned}
 \Delta E_0 = & -\frac{\hbar \omega_L}{2\pi} \int_0^\infty f d \ln |A(if)| \\
 = & -\frac{\hbar \omega_L}{2\pi} \int_0^\infty f d \ln \{1 + (P-1)(\sqrt{1+f^2} - f)^2\} \\
 & -\frac{\hbar \omega_L}{2\pi} \int_0^\infty f d \ln \left\{ 1 + \frac{Q}{P} (P-1) \frac{f(\sqrt{1+f^2} - f)^2}{(1+f^2)^{1/2}} \right. \\
 & \left. + \left( \frac{Q}{P} - 1 \right) \frac{f}{(1+f^2)^{1/2}} \right\} \quad (3.3)
 \end{aligned}$$

where we have put  $P = \gamma'/\gamma$  and  $Q = M'/M$  and have taken the  $D$ -shaped contour describ-

ed in Section II as our path of integration. These integrals have been evaluated for the conditions  $P > \frac{4}{Q} \left( 2 - \frac{P}{Q} - P \right) > 0$ ,  $Q > 0$ , with the result that

$$\begin{aligned} \Delta E_0 = & \frac{\hbar\omega_L}{2\pi} \left\{ \frac{P}{2\sqrt{P-1}} \cos^{-1} \left( \frac{2-P}{P} \right) - 1 \right\} \\ & + \frac{\hbar\omega_L}{2\pi} \left\{ 1 - \frac{\pi}{2} + \frac{A-B\alpha}{(\beta-\alpha)(\alpha-1)} + \frac{A-B\beta}{(\alpha-\beta)(\beta-1)} \right. \\ & + \frac{\alpha(A-B\alpha)}{(\beta-\alpha)(1-\alpha)} \frac{1}{\sqrt{1-\alpha^2}} \ln \left( \frac{1+\sqrt{1-\alpha^2}}{\alpha} \right) \\ & \left. + \frac{\beta(A-B\beta)}{(\alpha-\beta)(1-\beta)} \frac{1}{\sqrt{1-\beta^2}} \ln \left( \frac{1+\sqrt{1-\beta^2}}{\beta} \right) \right\} \end{aligned} \quad (3.4)$$

where

$$\begin{aligned} A = & \frac{PQ-2P}{2Q-P-PQ} \quad \alpha = \frac{P + \sqrt{P^2 - 4P \left( 2 - \frac{P}{Q} - P \right)}}{2 \left( 2 - \frac{P}{Q} - P \right)} \\ B = & \frac{4Q-2P-3PQ}{2Q-P-PQ} \quad \beta = \frac{P - \sqrt{P^2 - 4P \left( 2 - \frac{P}{Q} - P \right)}}{2 \left( 2 - \frac{P}{Q} - P \right)} \end{aligned}$$

The function  $\Omega(f) = \frac{d}{df} \ln |J(if)|$  is readily evaluated from (3.2) in the two limiting cases of small  $f$  and large  $f$  with the results that

$$\Omega(f) \xrightarrow{f \rightarrow 0} \frac{1}{P} (PQ - 3P + 2) + O(f^2) \quad (3.5a)$$

$$\Omega(f) \xrightarrow{f \rightarrow \infty} \frac{2}{f} \left\{ - \left( \frac{PQ-2Q}{2Q} \right) \frac{1}{f^2} + \dots \right\}. \quad (3.5b)$$

These expressions in conjunction with eqs. (2.20) and (2.26) lead immediately to the following low and high temperature expansions for the change in free energy:

$$JA(T)_{L.T.} = JE_0 - \frac{\kappa T}{\pi} \left\{ \frac{\pi^2}{6} \frac{PQ-3P+2}{P} \left( \frac{\kappa T}{\hbar\omega_L} \right) + \dots \right\} \quad (3.6a)$$

$$\begin{aligned} JA(T)_{H.T.} = & \frac{1}{2} \kappa T \ln \frac{P^2}{Q} + \kappa T \left\{ \frac{1}{24} \left[ \frac{P}{2} \left( \frac{Q+1}{Q} \right) - 1 \right] \left( \frac{\hbar\omega_L}{\kappa T} \right)^2 \right. \\ & \left. - \frac{1}{2880} \left[ \frac{P^2}{4} \left( \frac{Q+1}{Q^2} \right) + \frac{1}{8} (1+P)^2 - 1 \right] \left( \frac{\hbar\omega_L}{\kappa T} \right)^4 + \dots \right\}. \end{aligned} \quad (3.6b)$$



(ii) *An Isotopic Impurity*

An isotope impurity is one which alters only the mass of the atom at a particular lattice point and leaves the force constants unchanged. It is the special case of the general defect considered in (i) which results from putting  $P=\gamma'/\gamma=1$ . The expressions for the changes in zero-point energy and free energy follow from eqs. (3.4) and (3.6) on making this substitution with the results that

$$\Delta E_0 = \frac{\hbar\omega_L}{2} \left\{ (1-\varepsilon^2)^{-1/2} \cos^{-1}(-\varepsilon) - \frac{\pi}{2} \right\} \quad (3.7)$$

$$\Delta A(T)_{L,T} = \Delta E_0 + \frac{\varepsilon\kappa T}{\pi} \left\{ \frac{\pi^2}{6} \left( \frac{\kappa T}{\hbar\omega_L} \right) + \frac{\pi^4}{90} (3-2\varepsilon^2) \left( \frac{\kappa T}{\hbar\omega_L} \right)^3 \dots \right\} \quad (3.8a)$$

$$\begin{aligned} \Delta A(T)_{H,T} = & -\frac{\kappa T}{2} \ln(1-\varepsilon) + \kappa T \left\{ \frac{1}{24} \left[ \frac{1}{2} \left( \frac{\varepsilon}{1-\varepsilon} \right) \left( \frac{\hbar\omega_L}{\kappa T} \right)^2 \right. \right. \\ & \left. \left. - \frac{1}{2880} \left[ \frac{1}{2} \left( \frac{2-\varepsilon}{(1-\varepsilon)^2} \right) - \frac{1}{2} \right] \left( \frac{\hbar\omega_L}{\kappa T} \right)^4 \dots \right] \right\} \end{aligned} \quad (3.8b)$$

where for simplicity we have put  $\varepsilon=1-Q$ .

However, the principal reason for discussing this case separately is that an alternative expansion of the temperature dependent part of the free energy can be obtained in the following way.

The expression for  $\Delta I_n$ , eq. (2.18), becomes in this case

$$\Delta I_n = \frac{1}{\pi} \int_0^\infty \sin \alpha_n f \frac{df}{df} \ln [1 - \varepsilon f(1+f^2)^{-1/2}] df \quad (3.9)$$

which for small  $\varepsilon$  ( $|\varepsilon| < 1$ ) can be expanded as

$$\begin{aligned} \Delta I_n = & -\frac{\varepsilon}{\pi} \int_0^\infty \frac{\sin \alpha_n f}{(1+f^2)^{3/2}} df - \frac{\varepsilon^2}{\pi} \int_0^\infty \frac{f \sin \alpha_n f}{(1+f^2)^2} df + O(\varepsilon^3) \\ = & -\frac{\varepsilon \alpha_n}{\pi} \int_0^{\pi/2} e^{-\alpha_n \cos \theta} \cos \theta d\theta - \frac{\varepsilon^2}{4} \alpha_n e^{-\alpha_n} + O(\varepsilon^3). \end{aligned}$$

If we multiply this result by  $-\kappa T/n$  and sum from  $n=1$  to  $\infty$  according to eq. (2.26), we obtain

$$\begin{aligned} \Delta A(T) = & \Delta E_0 + \frac{\varepsilon \hbar\omega_L}{\pi} \int_0^{\pi/2} \frac{\cos \theta d\theta}{e^{\hbar\omega_L \cos \theta / \kappa T} - 1} \\ & + \varepsilon^2 \frac{\hbar\omega_L}{4} \frac{1}{e^{\hbar\omega_L / \kappa T} - 1} + O(\varepsilon^3), \end{aligned} \quad (3.10)$$

an expansion which is valid over the entire temperature range. The integral appearing in eq. (3.10) can be evaluated as a series expansion in powers of  $T$  or  $T^{-1}$ , or, just as easily, by numerical integration.

 (iii) *A Pair of Isotope Impurities*

If two like isotope defects with masses  $M' = (1 - \varepsilon)M$  are separated by  $m$  lattice spacings in a linear chain the defect determinant  $|\mathcal{A}(\omega)|$  is found to be

$$|\mathcal{A}(\omega)| = 1 - \varepsilon^2 M^2 \omega^4 \frac{g^2(m)}{[1 + \varepsilon M \omega^2 g(0)]^2} \\ \simeq 1 - \varepsilon^2 \frac{f^2}{1 + f^2} (\sqrt{1 + f^2} - f)^{4|m|} + O(\varepsilon^3) \quad (3.11)$$

in the weak defect limit,  $|\varepsilon| \ll 1$ . The change in zero point energy to this approximation is given by

$$\Delta E_0 = -\varepsilon^2 \frac{\hbar \omega_L}{2\pi} \int_0^\infty \frac{f^2 (\sqrt{1 + f^2} - f)^{4|m|}}{1 + f^2} df \\ = -\varepsilon^2 \frac{\hbar \omega_L}{4\pi} \left\{ \frac{8|m|}{16m^2 - 1} - \Psi\left(\frac{3}{4} + |m|\right) + \Psi\left(\frac{1}{4} + |m|\right) \right\} \quad (3.12)$$

where

$$\Psi(z) = \frac{d}{dz} \ln \Gamma(z).$$

The temperature dependent part of the interaction free energy can readily be obtained using the methods of Section II. In the weak defect limit  $\mathcal{Q}(f)$  becomes

$$\mathcal{Q}(f) = -\varepsilon^2 M^2 \omega_L^4 \frac{d}{df} [f^2 g^2(m; if)] \\ = -\varepsilon^2 \left\{ \frac{(\sqrt{1 + f^2} - f)^{4|m|}}{(1 + f^2)^2} - (4|m|f^3 + 2f) \right. \\ \left. - \frac{4|m|f^2}{(1 - f^2)^2} (\sqrt{1 + f^2} - f)^{4|m|} \right\}. \quad (3.13)$$

Evaluating the small  $f$  expansion of  $\mathcal{Q}(f)$  we obtain for the low temperature expansion of the interaction free energy (to  $O(\varepsilon^2)$ )

$$\Delta A(T) = \Delta E_0 - \frac{\kappa T}{\pi} \varepsilon^2 \left\{ 24|m| \frac{\pi^4}{90} \left( \frac{\kappa T}{\hbar \omega_L} \right)^3 \right. \\ \left. + \frac{16\pi^6}{189} (16|m|^3 + 24m^2 + |m|) \left( \frac{\kappa T}{\hbar \omega_L} \right)^5 \dots \right\}. \quad (3.14)$$

This type of expansion is mainly useful for small separations between defects. For large  $|m|$  we can use the asymptotic form for the Green's function, eq.

$$g(m; if) \sim -\frac{1}{4if} e^{-2|m|f},$$

so that here

$$\Delta I_n \simeq -\frac{\varepsilon^2}{\pi} \int_0^\infty \sin \alpha_n f d\{f^2 e^{-4|m|f}\}$$

$$= 128\alpha_n \frac{\varepsilon^2}{\pi} \left\{ \frac{|m|^3}{(\alpha_n^2 + 16m^2)^3} - \frac{3}{16} \alpha_n^2 \frac{|m|}{(\alpha_n^2 + 16m^2)^2} \right\}. \quad (3.15)$$

Multiplying this expression by  $-\frac{\kappa T}{N}$  and summing over  $n$  from 1 to  $\infty$  we obtain finally

$$\begin{aligned} \Delta A(T) = \Delta E_0 - \frac{128\varepsilon^2}{\pi} \frac{\hbar\omega_L}{\hbar\omega_L} \left( \frac{\kappa T}{\hbar\omega_L} \right)^6 \left\{ 4|m|^3 F_1 \left( \frac{4m\kappa T}{\hbar\omega_L} \right) \right. \\ \left. - \frac{3}{16} |m| \left( \frac{\hbar\omega_L}{\kappa T} \right)^2 F_2 \left( \frac{4m\kappa T}{\hbar\omega_L} \right) \right\} \end{aligned} \quad (3.16)$$

where

$$\begin{aligned} F_1(x) = \frac{1}{2} \left\{ \frac{3\pi}{8x^5} + \frac{\pi^3}{4x^3} \frac{\cosh \pi x}{\sinh^3 \pi x} + \frac{3\pi^2}{8x^4} \frac{1}{\sinh^2 \pi x} \right. \\ \left. + \frac{3\pi}{8x^5} \frac{\cosh \pi x - \sinh \pi x}{\sinh \pi x} - \frac{1}{x^6} \right\} \end{aligned} \quad (3.17a)$$

$$F_2(x) = \frac{1}{2} \left\{ \frac{\pi}{2x^3} + \frac{\pi^2}{2x^2} \frac{1}{\sinh^2 \pi x} + \frac{\pi}{2x^3} \frac{\cosh \pi x - \sinh \pi x}{\sinh \pi x} - \frac{1}{x^4} \right\}. \quad (3.17b)$$

At high temperatures we need not use the moment method in this case as another method becomes available to us. In the weak defect limit

$$\begin{aligned} \Delta I_n = - \frac{\varepsilon^2 M^2 \omega_L^4}{\pi} \int_0^\infty \sin \alpha_n f \frac{d}{df} \{ f^4 g^2(m; if) \} df \\ = \alpha_n \frac{\varepsilon^2 M^2 \omega_L^4}{\pi} \int_0^\infty f^4 g^2(m; if) \cos \alpha_n f df. \end{aligned} \quad (3.18)$$

If we multiply this by  $-\frac{\kappa T}{n}$  and interchange the order of summation and integration

$$\begin{aligned} \Delta A(T) = \Delta E_0 - \kappa T \frac{\varepsilon^2 M^2 \omega_L^4}{\pi} \int_0^\infty \left\{ \sum_{n=1}^\infty \frac{\alpha_n}{n} \cos \alpha_n f \right\} f^4 g^2(m; if) df \\ = \Delta E_0 - \frac{\hbar\omega_L \varepsilon^2}{\pi} \int_0^\infty \left\{ \pi \sum_j \delta \left( \frac{\hbar\omega_L}{\kappa T} f - 2\pi j \right) - \frac{1}{2} \right\} \frac{f^2}{1+f^2} (1+f^2-f)^{4|m|} df, \end{aligned} \quad (3.19)$$

where we have used the relation

$$\sum_{n=1}^\infty \cos nx = \pi \sum_j \delta(x - 2\pi j) - \frac{1}{2}. \quad (3.20)$$

The  $\delta$ -function converts the integral into a sum with the result that

$$\Delta A(T) = -\varepsilon^2 \kappa T \sum_{j=1}^\infty \frac{\xi^2 j^2}{1 - \xi^2 j^2} [V' 1 + \xi^2 j^2 - \xi j]^{4|m|} \quad (3.21)$$

where

$$z = \frac{2\pi kT}{\hbar\omega_L}.$$

For  $T > \frac{\hbar\omega_L}{2\pi k}$  the sum can be expanded in inverse powers of  $T$ :

$$\begin{aligned} \Delta A(T) = -\varepsilon^2 \kappa T \left( \frac{\hbar\omega_L}{\kappa T} \right)^{|m|} & \left\{ \frac{B_{2m}}{2^{4m+1} (4m)!} - \frac{(m+1) B_{2m+1}}{2^{4m+1} (4m+2)!} \left( \frac{\hbar\omega_L}{\kappa T} \right)^3 \right. \\ & \left. + \frac{4m^2 + 11m + 8}{8} \frac{\beta_{2m+2}}{2^{4m+1} (4m+4)} \left( \frac{\hbar\omega_L}{\kappa T} \right)^4 \dots \right\}. \quad (3.22) \end{aligned}$$

Thus in the weak defect limit the first non-vanishing term in the expansion of the interaction free energy in inverse powers of temperature depends on the separation between the defects. This result can also be established in the general case of  $\varepsilon = 1$  either by the method used to obtain eq. (3.21) or through the use of the moment-difference generating property of  $\mathcal{Q}(f)$ . This qualitative feature of the high temperature interaction free energy had been noted previously by Montroll, Maradudin, and Weiss.

#### (iv) Anomalous Force Constants

The next defect we consider is a single anomalous force constant which is the result of replacing the force constant  $\gamma$  for the interaction between two neighboring atoms by a different force constant  $\gamma''$ .

Anomalous force constants have some features in common with vacancies in the lattice. We shall first consider the case in which there is a single such defect in a monatomic linear chain.

The defect determinant in this case is easily shown to be

$$|\mathcal{A}(\omega)| = 1 - 2\gamma\delta[g(0) - g(1)] \quad (3.23)$$

where  $\delta = \left( \frac{\gamma''}{\gamma} - 1 \right)$ . The change in zero point energy is given by

$$\begin{aligned} \Delta E_0 &= -\frac{\hbar\omega_L}{2\pi} \int_0^\infty f d \ln \left[ 1 - \delta \frac{(f - \sqrt{1+f^2})}{\sqrt{1+f^2}} \right] \\ &= \frac{\hbar\omega_L}{2\pi} \left[ \frac{1+\delta}{\sqrt{1+2\delta}} \cos^{-1} \left( \frac{-\delta}{1+\delta} \right) - \frac{\pi}{2} \right] \end{aligned} \quad (3.24)$$

where  $\delta$  is taken positive, meaning thereby that the anomalous force constant  $\gamma''$  is stiffer. From (2.28)

$$\begin{aligned} \Delta I_n &= \frac{1}{\pi} \int_0^\infty \sin \alpha_n f \frac{d}{df} \ln \left[ 1 - \delta \frac{(f - \sqrt{f^2+1})}{\sqrt{f^2+1}} \right] df \\ &= -\frac{\delta}{\pi} \int_0^\infty \frac{\sin \alpha_n f}{1+f^2} \frac{df}{\sqrt{1+f^2+\delta} (\sqrt{1+f^2}-f)}. \end{aligned} \quad (3.25)$$

This integral can be evaluated in a power series of  $\delta$  when  $\delta$  is small. For this case

$$\begin{aligned} \Delta I_n &= -\frac{\delta}{\pi} \int_0^\infty \frac{\sin \alpha_n f}{(1+f^2)^{3/2}} df + \frac{\delta^2}{\pi} \left[ \int_0^\infty \frac{\sin \alpha_n f}{(1+f^2)^{3/2}} df - \int_0^\infty \frac{f \sin \alpha_n f}{(1+f^2)^2} df \right] \\ &\quad + O(\delta^3) \\ &= -\frac{\delta}{\pi} \alpha_n \int_0^{\pi/2} e^{-\alpha_n \cos \theta} \cos \theta d\theta + \frac{\delta^2}{\pi} \left[ \int_0^{\pi/2} e^{-\alpha_n \cos \theta} \cos \theta d\theta - \frac{\pi}{4} \alpha_n e^{-\alpha_n} \right] \\ &\quad + O(\delta^3). \end{aligned}$$

Multiplying this by  $-\frac{\kappa T}{n}$  and summing from  $n=1$  to  $\infty$ , we get

$$\begin{aligned} \Delta A(T) &= \frac{\hbar \omega_L}{2\pi} \left[ \frac{1+\delta}{\sqrt{1+2\delta}} \cos^{-1} \left( \frac{-\delta}{1+\delta} \right) - \frac{\pi}{2} \right] \\ &\quad + \frac{\delta}{\pi} \hbar \omega_L \int_0^{\pi/2} \frac{\cos \theta}{e^{\hbar \omega_L / \kappa T \cos \theta} - 1} d\theta - \frac{\delta^2}{\pi} \hbar \omega_L \left[ \int_0^{\pi/2} \frac{\cos \theta}{e^{\hbar \omega_L / \kappa T \cos \theta} - 1} d\theta \right. \\ &\quad \left. - \frac{\pi}{4} \frac{1}{e^{\hbar \omega_L / \kappa T} - 1} \right] + O(\delta^3). \end{aligned} \quad (3.26)$$

The high and low temperature expansions are obtained without much difficulty, and they are given by

$$\begin{aligned} \Delta A(T)_{L.T.} &= \frac{\hbar \omega_L}{2\pi} \left[ \frac{1+\delta}{\sqrt{1+2\delta}} \cos^{-1} \left( \frac{-\delta}{1+\delta} \right) - \frac{\pi}{2} \right] \\ &\quad + \frac{\kappa T}{\pi} \left\{ \frac{\pi^2}{6} \left( \frac{\delta}{1+\delta} \right) \left( \frac{\kappa T}{\hbar \omega_L} \right) + \frac{\pi^4}{45} \left( \frac{\delta}{1+\delta} \right) \left[ \frac{3}{2} - \left( \frac{\delta}{1+\delta} \right)^2 \right] \left( \frac{\kappa T}{\hbar \omega_L} \right)^3 + \dots \right\} \end{aligned} \quad (3.27a)$$

$$\Delta A(T)_{H.T.} = \frac{1}{2} \kappa T \ln(1+\delta) + \kappa T \left\{ \frac{\delta}{48} \left( \frac{\hbar \omega_L}{\kappa T} \right)^2 - \frac{(3+\delta)\delta}{11520} \left( \frac{\hbar \omega_L}{\kappa T} \right)^4 + \dots \right\}. \quad (3.27b)$$

A more interesting case is that of two interacting defects of this type.

The correction determinant for this case is

$$\begin{aligned} |\Delta(\omega)| &= 1 + \delta^2 M \omega^2 \gamma \frac{g(m)[g(m-1) - g(m+1)]}{\{1 - 2\gamma\delta[g(0) - g(1)]\}^2} \\ &\cong 1 + \delta^2 M \omega^2 \gamma g(m)[g(m-1) - g(m+1)] \end{aligned} \quad (3.28)$$

when  $\delta$  is small. Here  $m$  is the number of lattice spacings between the defects. The expression for the zero point interaction energy is (for  $\delta$  small),

$$\begin{aligned} \Delta E_0 &= -\frac{\hbar \omega_L}{2\pi} \int_0^\infty f d \ln \left[ 1 - \delta^2 \frac{f}{\sqrt{1+f^2}} (\sqrt{1+f^2} - f)^{4|m|} \right] \\ &\cong -\frac{\hbar \omega_L}{2\pi} \delta^2 \int_0^\infty \frac{f}{\sqrt{1+f^2}} (\sqrt{1+f^2} - f)^{4|m|} df \end{aligned} \quad (3.29)$$



$$= -\frac{\hbar\omega_L}{2\pi} \frac{\delta^2}{\delta^2} \frac{1}{16m^2-1}.$$

Proceeding in the same way as we did to obtain equation (3.21) we get the following expression for the interaction free energy :

$$\Delta A(T) = -\delta^2 \kappa T \left\{ \sum_{j=0}^{\infty} \frac{\zeta^j}{\sqrt{1+\zeta^2 j^2}} [\sqrt{1+\zeta^2 j^2} - \zeta j]^{4|m|} \right\}. \quad (3.30)$$

The low and high temperature expansions of the interaction free energy are (in the weak defect limit)

$$\Delta A(T)_{L.T.} = \Delta E_0 + \frac{\kappa T}{\pi} \left\{ 2m \delta^2 \frac{\pi^4}{45} \left( \frac{\kappa T}{\hbar\omega_L} \right)^3 + \dots \right\} \quad (3.31)$$

and

$$\begin{aligned} \Delta A(T)_{H.T.} = & -\delta^2 \kappa T \left( \frac{\hbar\omega_L}{2\kappa T} \right)^{4|m|} \left\{ \frac{B_{2m}}{(4m)!} - \frac{(2m+1)}{2} \frac{B_{2m+1}}{(4m+2)!} \left( \frac{\hbar\omega_L}{2\kappa T} \right)^2 \right. \\ & \left. + \frac{(6m+1)}{4} \frac{B_{2m-2}}{(4m+4)!} \left( \frac{\hbar\omega_L}{2\kappa T} \right)^4 - \dots \right\} \end{aligned} \quad (3.32)$$

where the  $B_n$  are Bernoulli numbers.

Again it should be emphasized that it is not necessary to restrict the calculation of interaction energies to the weak defect limit. The methods described here are completely general.

#### (v) The Lattice Vacancy

We can apply the results obtained for the anomalous force constants to a calculation to the self free energy of a lattice vacancy. We proceed in the following way. A vacancy can be characterized by setting the defect mass  $M'$  in eq. (3.2) equal to zero while at the same time stiffening the force constant  $\gamma'$  associated with its interactions with its nearest neighbors. This choice for  $M'$  and  $\gamma'$  resembles a vacancy in a crystal lattice if one examines what happens when an atom is removed from a lattice site in a monatomic lattice. The repulsive force which kept its neighbors in their places is removed so that a tendency exists for them to fall into the hole. We characterize this tendency by coupling the neighbors of the hole to the hole's lattice point by a stronger than normal spring ( $\gamma' > \gamma$ ), or by an alternative model in which the neighbors on opposite sides of the hole are coupled to gether with a spring constant  $\gamma'' = \frac{1}{2} \gamma' > \gamma$ .

If we set  $M'$  equal to zero in eq. (3.2) and denote the defect force constant by  $\gamma'$  the equation for the impurity frequency,  $|\Delta(\omega)|=0$ , is

$$(\gamma' - \gamma)[g(0) - g(2)] = 1 \quad (3.33)$$

which is readily solved with the aid of eq. (2.14b) to give the result that

$$\omega_{vac.}^2 = \omega_L^2 \frac{P^2}{4(P-1)} \quad (3.34)$$

where  $P = (\gamma'/\gamma) \geq 2$ .

The contribution to the change in free energy from this impurity frequency is

$$\Delta A(T) = \frac{1}{2} \hbar \omega_L \left[ \frac{P}{2\sqrt{P-1}} - 1 \right] + \kappa T \ln \frac{1 - e^{-\frac{\hbar \omega_L}{\kappa T} \frac{P}{2\sqrt{P-1}}}}{1 - e^{-\frac{\hbar \omega_L}{\kappa T}}} \quad (3.35)$$

The contribution to the change in free energy  $\Delta A_b$  arising from the small shifts of the in-band frequencies can be obtained as follows:

(a) We insert an anomalous bond with force constant  $\gamma''$  into the lattice. The change in free energy  $\Delta A_b$  arising from the in-band frequency shifts is the difference between the total self free energy  $\Delta A_{a.f.c.}$  given by eq. (3.27) and the contribution from the localized mode whose frequency is given by the solution of

$$2(\gamma'' - \gamma)[g(0) - g(1)] = 1. \quad (3.36)$$

This impurity frequency is found to be

$$\omega_{a.f.c.}^2 = \omega_L^2 \frac{(P')^2}{2P' - 1} \quad (3.37)$$

where  $P' = (\gamma''/\gamma)$ . Thus we find that

$$\begin{aligned} \Delta A_b^{(1)}(T) = \Delta A(T)_{a.f.c.} - \frac{1}{2} \hbar \omega_L \left[ \frac{P'}{\sqrt{2P'-1}} - 1 \right] \\ - \kappa T \ln \frac{1 - e^{-\frac{\hbar \omega_L}{\kappa T} \frac{P'}{\sqrt{2P'-1}}}}{1 - e^{-\frac{\hbar \omega_L}{\kappa T}}} \end{aligned} \quad (3.38)$$

(b) We now remove one particle from the chain with

$$\Delta A_b^{(2)}(T) = -\frac{\hbar \omega_L}{\pi} - \frac{2\kappa T}{\pi} \int_0^{\pi/2} \ln \left( 1 - e^{-\frac{\hbar \omega_L}{\kappa T} \sin \theta} \right) d\theta, \quad (3.39)$$

since this is the total free energy per particle in a long chain.

(c) Without changing the topology of the lattice the anomalous bond is chosen to span the vacancy so that it is extended over two lattice spacings rather than the usual one per bond. The choice  $P' = \frac{1}{2}P$  is made to correspond to a consistent model of a vacancy. The total self free energy of a vacancy in a linear chain is then given by the sum of (3.35), (3.38), and (3.39):

$$\Delta A(T)_{\text{vacancy}} = \Delta A(T)_{a.f.c.} \Big|_{P' = 1/2 P} - \frac{\hbar \omega_L}{\pi}$$

$$-\frac{2\kappa T}{\pi} \int_0^{\pi/2} \ln \left( 1 - e^{-\frac{\hbar\omega_L}{\kappa T} \sin \theta} \right) d\theta. \quad (3.40)$$

Low and high temperature expansions for this energy are given by

$$\begin{aligned} \Delta A(T)_{L.T.} = & \frac{\hbar\omega_L}{2\pi} \left[ \frac{P}{2\sqrt{P-1}} \cos^{-1} \left( \frac{2-P}{P} \right) - \frac{\pi}{2} - 2 \right] \\ & + \frac{\kappa T}{\pi} \left\{ -\frac{\pi^2}{6} \left( \frac{P+2}{P} \right) \left( \frac{\kappa T}{\hbar\omega_L} \right) + \frac{\pi^4}{45} \left[ \frac{P-2}{P} \left( \frac{3}{2} - \frac{(P-2)^2}{P^2} \right) - 1 \right] \left( \frac{\kappa T}{\hbar\omega_L} \right)^3 \dots \right\} \\ \Delta A(T)_{H.T.} = & \frac{1}{2} \kappa T \ln \frac{2P\kappa^2 T^2}{\hbar^2 \omega_L^2} + \kappa T \left\{ \frac{P-4}{96} \left( \frac{\hbar\omega_L}{\kappa T} \right)^2 \right. \end{aligned} \quad (3.41a)$$

$$\left. - \frac{P^2 + 2P - 14}{46080} \left( \frac{\hbar\omega_L}{\kappa T} \right)^4 + \dots \right\}. \quad (3.41b)$$

#### § 4. Defects in diatomic lattices

With the aid of the “ $M^*$ ” transformation and the expansions developed in Section II, the calculation of the effects on the thermodynamic properties of diatomic lattices becomes almost as easy as the corresponding calculations for monatomic lattices.

We consider first the case of a single defect which alters both the mass of a particle and the force constants associated with its interactions with its nearest neighbors. We assume that the particle at the origin whose mass was originally  $M$ , is replaced by a particle of mass  $M_1(1-\epsilon)$ . Furthermore, its force constant  $\gamma$  is changed to  $\gamma(1-\gamma)$ . We consider only the case  $M_1 > M_2$  since the discussion in the case  $M_1 < M_2$  follows along identical lines.

The equations for the time-independent parts of the displacement components are given below.

$$\begin{aligned} M_1(1-\epsilon)\omega^2 u(0) + \gamma(1-\gamma)[u(1) - 2u(0) + u(-1)] &= 0 \\ M_2\omega^2 u(1) + \gamma[u(2) - u(1)] - \gamma(1-\gamma)[u(1) - u(0)] &= 0 \\ M_2\omega^2 u(-1) + \gamma(1-\gamma)[u(0) - u(-1)] - \gamma[u(-1) - u(-2)] &= 0 \end{aligned} \quad (4.1)$$

We now go over into the “ $r$ ” notation described by Montroll and Potts.<sup>3</sup> The transformed equations are

$$\begin{aligned} M^*\omega^2 r(0) + \gamma[r(1) - 2r(0) + r(-1)] &= (M_1\epsilon\omega^2 - 2\gamma\gamma) \frac{(M_2\omega^2 - 2\gamma)^{1/2}}{M_1\omega^2 - 2\gamma} r(0) \\ M^*\omega^2 r(1) + \gamma[r(2) - 2r(1) + r(0)] &= \gamma\gamma \left\{ r(0) - \frac{(M_1\omega^2 - 2\gamma)^{1/2}}{(M_2\omega^2 - 2\gamma)^{1/2}} r(1) \right\} \\ M^*\omega^2 r(-1) + \gamma[r(0) - 2r(-1) + r(-2)] &= \gamma\gamma \left\{ r(0) - \frac{(M_1\omega^2 - 2\gamma)^{1/2}}{(M_2\omega^2 - 2\gamma)^{1/2}} r(-1) \right\} \end{aligned} \quad (4.2)$$

so that the determinant  $|J(\omega)| = |I + M_0^{-1}(\omega) \partial M(\omega)|$  becomes

$$\begin{vmatrix} 1+g(0)\epsilon_1-g(1)\epsilon_2 & -g(0)\epsilon_2+g(1)\epsilon_3-g(2)\epsilon_2 & -g(1)\epsilon_2+g(2)\epsilon_1 \\ g(-1)\epsilon_1-g(0)\epsilon_2 & 1-g(-1)\epsilon_2+g(0)\epsilon_3-g(1)\epsilon_2 & -g(0)\epsilon_2+g(1)\epsilon_1 \\ g(-2)\epsilon_1-g(-1)\epsilon_2 & -g(-2)\epsilon_2+g(-1)\epsilon_3-g(0)\epsilon_2 & 1-g(-1)\epsilon_2+g(0)\epsilon_1 \end{vmatrix} \quad (4.3)$$

where

$$\begin{aligned} \epsilon_1 &= \gamma\eta \frac{(M_1\omega^2 - 2\gamma)^{1/2}}{(M_2\omega^2 - 2\gamma)^{1/2}} \\ \epsilon_2 &= \gamma\eta \\ \epsilon_3 &= (2\gamma\eta - M_1\omega^2\epsilon) \frac{(M_2\omega^2 - 2\gamma)^{1/2}}{(M_1\omega^2 - 2\gamma)^{1/2}} \end{aligned} \quad (4.4)$$

This determinant factors as follows :

$$\begin{aligned} |J(\omega)| &= \{1+g(0)\epsilon_1-g(2)\epsilon_1\} \{ (1+g(0)\epsilon_3-2g(1)\epsilon_2) (1+g(0)\epsilon_1 \\ &\quad -2g(1)\epsilon_2+g(2)\epsilon_1) -2(g(1)\epsilon_1-g(0)\epsilon_2) (g(2)\epsilon_2-g(1)\epsilon_3+g(0)\epsilon_2) \}, \end{aligned} \quad (4.5)$$

In the special case of an isotope defect ( $\epsilon_1=\epsilon_2=0$ ,  $\epsilon_3=-M_1\epsilon\omega^2 \frac{(M_2\omega^2-2\gamma)^{1/2}}{(M_1\omega^2-2\gamma)^{1/2}}$ ) this reduces to

$$|J(\omega)| = 1 - M_1\epsilon\omega^2 \frac{(M_2\omega^2-2\gamma)^{1/2}}{(M_1\omega^2-2\gamma)^{1/2}} g(0). \quad (4.6)$$

The use of (4.5) for finding changes in thermodynamic quantities leads to cumbersome integrals. The essential features are there in the isotope case ; so we will consider the isotope case alone and use (4.6).

For the evaluation of the low and high temperature expansions for the changes in thermodynamic properties due to the presence of the defect we need to evaluate the small  $f$  and large  $f$  expansions of the function

$$\Omega(f) = \frac{d}{df} \ln |J(if)|$$

respectively with  $f = \frac{\omega}{\omega_L}$ . Using the expressions for the  $g(k)$  given in eq. (2.15) we obtain for the leading terms

$$\begin{aligned} \Omega(f) \xrightarrow{f \rightarrow 0} & \left\{ -\epsilon \frac{\omega_2}{\omega_1} - \epsilon^2 \frac{\omega_2^2}{\omega_1^2} f^2 \right. \\ & \left. - \left[ \epsilon^3 \frac{\omega_2^3}{\omega_1^3} - \frac{3}{2} \epsilon \frac{\omega_2}{\omega_1} \left( \frac{\omega_1^2 \omega_2^2 - \omega_1^2 \omega_L^2 + \omega_2^2 \omega_L^2}{\omega_1^2 \omega_2^2} \right) \right] f^4 + \dots \right\} \end{aligned} \quad (4.7)$$

$$\Omega(f) \xrightarrow{f \rightarrow \infty} \frac{2}{f} \left\{ -\frac{\epsilon}{1-\epsilon} \frac{A}{f^2} + \left( \frac{\epsilon^2}{1-\epsilon^2} B - \frac{2\epsilon}{1-\epsilon} C \right) \frac{1}{f^4} + \dots \right\} \quad (4.8)$$

where

$$A = \frac{1}{2} - \frac{\alpha^2}{2} - \frac{\beta^2}{2}, \quad \alpha = \frac{\omega_1}{\omega_I}, \quad \beta = \frac{\omega_2}{\omega_I}$$

$$B = \frac{1}{4} (1 + \alpha^2)^2 + \frac{\beta^2}{4} (\beta^2 - 2\alpha^2 - 2)$$

$$C = \frac{1}{8} \left[ (\beta^2 + 3\alpha^2) (\beta^2 - \alpha^2) + 2\beta^2 - 2\alpha^2 - 3 \right].$$

We can immediately write down for the leading terms in the low and high temperature expansions for the change in free energy:

$$(\Delta A)_{L.T.} = \Delta E_0 + \frac{\kappa T}{\pi} \left\{ \frac{\pi^2 \epsilon \omega_2}{6 \omega_1} \left( \frac{\kappa T}{\hbar \omega_L} \right) + \frac{\pi^4 \omega_2^2 \epsilon^2}{45 \omega_1^2} \left( \frac{\kappa T}{\hbar \omega_L} \right)^2 + \dots \right\} \quad (4.9)$$

$$(\Delta A)_{H.T.} = -\frac{\kappa T}{2} \ln(1 - \epsilon)$$

$$- \kappa T \left[ \frac{\epsilon A}{24(1 - \epsilon)} \left( \frac{\hbar \omega_L}{\kappa T} \right)^2 - \frac{1}{2880} \left( \frac{\epsilon^2 B}{1 - \epsilon} - \frac{2\epsilon^2 C}{1 - \epsilon} \right) \left( \frac{\hbar \omega_L}{\kappa T} \right)^4 + \dots \right]. \quad (4.10)$$

It remains only to evaluate the change in zero-point energy,  $\Delta E_0$ , which is given formally by equations (2.18) and (2.15) as

$$\Delta E_0 = -\frac{\hbar}{2\pi} \int_0^\infty \omega d \ln \left[ 1 - \frac{\epsilon f}{\sqrt{1 + f^2}} \left( \frac{f^2 + \beta^2}{f^2 + \alpha^2} \right)^{1/2} \right]. \quad (4.11)$$

This integral has been evaluated by Mazur, Montroll and Potts<sup>2</sup> and by Montroll and Potts<sup>3</sup> in two limiting cases,  $M_1 = \frac{1}{\zeta^2} M_2$ ,  $\zeta \ll 1$ , and  $M_1 = (1 + \gamma) M_2$ ,  $\gamma \ll 1$ . To terms linear in  $\zeta$  and  $\gamma$ , the results are

$$\Delta E_0 = \frac{1}{2} \hbar \omega_L \left\{ \frac{\pi}{\zeta} \left[ -\frac{1}{2} + \frac{1}{\pi} (1 - \epsilon^2)^{-1/2} \left( \frac{\pi}{2} + \sin^{-1} \epsilon \right) \right] + O(\zeta^2) + \dots \right\} \quad (4.12)$$

$$\Delta E_0 = \frac{1}{2} \hbar \omega_L \left\{ -\frac{1}{2} + \frac{1}{2\pi} (1 - \epsilon^2)^{-1/2} (\pi + 2 \sin^{-1} \epsilon) \right.$$

$$\left. + \frac{\epsilon \gamma}{8\pi(1 + \epsilon^2)} [2\sqrt{2} \ln(1 + \sqrt{2}) - \sqrt{2} \epsilon \pi + \frac{2\epsilon}{(1 - \epsilon^2)^{1/2}} (\pi + 2 \sin^{-1} \epsilon)] + \dots \right\}. \quad (4.13)$$

The more interesting case of a defect pair can be treated in the same manner as the monatomic case. We shall consider here the case of isotope defects only. There are three distinct possibilities:

- Both the defect atoms are on heavy sites,  $M'_1 = M_1(1 - \epsilon_1)$
- Both are on light sites,  $M'_2 = M_2(1 - \epsilon_2)$
- One is on a heavy site, and one on a light site.



As in the monatomic case, only terms up to  $\epsilon^2$  can be evaluated without difficulty so that the treatment we give here is valid for  $\epsilon$  small.

(a) Defect pair on heavy sites :

The correction determinant in this case is easily shown to be

$$|\mathcal{A}(i\omega)| \simeq 1 - \epsilon_1^2 \omega^4 M_1 M_2 \left( \frac{\omega^2 + \omega_2^2}{\omega^2 + \omega_1^2} \right) g(m, i\omega)^2 \quad (4.14)$$

where  $g$  is given by (2.15) and  $m$  is the number of lattice spacings between the defects.

The expression for the interaction energy is

$$\begin{aligned} \Delta E_0 &= -\frac{\hbar}{4\pi} \int_{-\infty}^{\infty} z d \ln \left[ 1 - \epsilon_1^2 z^4 M_1 M_2 \left( \frac{z^2 + \omega_2^2}{z^2 + \omega_1^2} \right) g(m)^2 \right] \\ &\simeq -\frac{\epsilon_1^2 M_1 M_2 \hbar}{2\pi} \int_0^{\infty} z^4 \left( \frac{z^2 + \omega_2^2}{z^2 + \omega_1^2} \right) g(m)^2 dz \end{aligned} \quad (4.15)$$

where we have integrated by parts and retained terms up to  $\epsilon_1^2$ . The integral can be evaluated with some approximations<sup>3)</sup> and the value is

$$\Delta E_0 \simeq -\frac{1}{m^3} \frac{\gamma^{1/2} \omega_2^2 \epsilon_1^2 \hbar M_1 M_2}{4\pi (M_1 + M_2)^{5/2} \omega_1^2} \quad (4.16)$$

The low and high temperature expansions of the interaction free energy are obtained as before, and are given by

$$\begin{aligned} (\mathcal{A})_{L.T.} &= \Delta E_0 - \frac{\kappa T}{\pi} \left\{ \frac{\pi^4}{45} \frac{\epsilon_1^2 \omega_2^2}{\omega_1^2} \left( \frac{\kappa T}{\hbar \omega_L} \right)^3 - \frac{\pi^6 \epsilon_1^2}{315 \omega_1^4} [(2m^2 - 1) \omega_L^4 + \omega_1^2 \omega_L^2 + \omega_1^4] \right. \\ &\quad \left. \times \left( \frac{\kappa T}{\hbar \omega_L} \right)^5 + \dots \right\} \end{aligned} \quad (4.17)$$

$$(\mathcal{A})_{H.T.} = -\kappa T \frac{\epsilon_1^2 (\omega_1 \omega_2)^{2m}}{2^{2m}} \left[ \frac{B_{2m}}{(4m)!} \left( \frac{\hbar}{\kappa T} \right)^{4m} - \frac{2B_{2m+1}}{(4m+2)!} \frac{\omega_1^2}{\omega_2^2} \left( \frac{\hbar}{\kappa T} \right)^{4m+2} + \dots \right] \quad (4.18)$$

(b) Defect Pair on light sites.

The correction determinant for this case is

$$|\mathcal{A}(i\omega)| \simeq 1 - \epsilon_2^2 M_1 M_2 \omega^4 \left( \frac{\omega^2 + \omega_1^2}{\omega^2 + \omega_2^2} \right) g(m, i\omega)^2 \quad (4.19)$$

The expression for the interaction energy

$$\begin{aligned} \Delta E_0 &= -\frac{\epsilon_2^2 M_1 M_2 \hbar}{4\pi} \int_{-\infty}^{\infty} z^4 \left( \frac{z^2 + \omega_1^2}{z^2 + \omega_2^2} \right) g^2(m) dz \\ &\simeq -\frac{1}{m^3} \frac{\gamma^{1/2} \omega_1^2 \epsilon_2^2 \hbar M_1 M_2}{4\pi (M_1 + M_2)^{5/2} \omega_2^2} \end{aligned} \quad (4.20)$$

The low and high temperature expansions of the interaction free energy are

$$(\mathcal{A})_{L.T.} = \Delta E_0 - \frac{\kappa T}{\pi} \left\{ \frac{\pi^4 \epsilon_2^2 \omega_1^2}{45 \omega_2^2} \left( \frac{\kappa T}{\hbar \omega_L} \right)^3 - \frac{\pi^6 \epsilon_2^2}{315 \omega_2^4} [(2m^2 - 1) \omega_L^4 + \omega_2^2 \omega_L^2 + \omega_2^4] \right\}$$

$$\times \left( \frac{\kappa T}{\hbar \omega_L} \right)^5 - \left\{ \quad \right\} \quad (4.21)$$

$$(JA)_{H.T.} = -\kappa T \frac{\epsilon_1^2 (\omega_1 \omega_2)^{2m}}{2^{2m}} \left[ \frac{B_m}{(4m)!} \left( \frac{\hbar}{\kappa T} \right)^{4m} - \frac{2B_{m-1} \omega_2^2}{(4m-2)!} \left( \frac{\hbar}{\kappa T} \right)^{4m-2} - \dots \right] \quad (4.22)$$

(c) Defects at heavy and light sites.

The correction determinant for this case is

$$|J(i\omega)| \cong 1 - \epsilon_1 \epsilon_2 M_1 M_2 \omega^4 g(m, i\omega)^2. \quad (4.23)$$

The expression for the interaction energy is

$$\begin{aligned} \Delta E_0 &\cong -\frac{\epsilon_1 \epsilon_2 M_1 M_2 \hbar}{2\pi} \int_0^\infty z^4 g^2(m) dz \\ &\cong -\frac{1}{m^3} \frac{\gamma^{1/2} \epsilon_1 \epsilon_2 \hbar M_1 M_2}{4\pi (M_1 + M_2)^{5/2}}. \end{aligned} \quad (4.24)$$

The low and high temperature expansions of the interaction free energy are

$$(JA)_{L.T.} = JE_0 - \frac{\kappa T}{\pi} \left\{ \frac{\pi^4 \epsilon_1 \epsilon_2}{45} \left( \frac{\kappa T}{\hbar \omega_L} \right)^3 - \frac{\pi^6 \epsilon_1 \epsilon_2}{315} \left[ \frac{2m^2 \omega_1^4}{\omega_1^2 \omega_2^2} - 1 \right] \left( \frac{\kappa T}{\hbar \omega_L} \right)^5 - \dots \right\} \quad (4.25)$$

$$(JA)_{H.T.} = -\frac{\kappa T \epsilon_1 \epsilon_2 (\omega_1 \omega_2)^{2m}}{2^{2m}} \left\{ \frac{B_m}{(4m)!} \left( \frac{\hbar}{\kappa T} \right)^{4m} - \frac{B_{m-1} \omega_1^2}{(4m-2)!} \left( \frac{\hbar}{\kappa T} \right)^{4m-2} - \dots \right\} \quad (4.26)$$

## § 5. The effects of a large number of impurities

Let us suppose that there are  $n$  defects in a lattice and that we are interested in  $\langle S(n) \rangle$ , the value of a thermodynamic function  $S$  averaged over all configurations of  $n$  defects in the lattice. By a well known formula from the calculus of finite differences  $\langle S(n) \rangle$  can be expressed as

$$\langle S(n) \rangle = \sum_{j=0}^n \binom{n}{j} \langle J^j S(0) \rangle \quad (5.1)$$

where

$$\langle JS(0) \rangle = \langle S(1) - S(0) \rangle \quad (5.2)$$

and

$$\langle J^{K+1} S(0) \rangle = \langle J(J^K S(0)) \rangle. \quad (5.3)$$

If we denote by  $W(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n)$  the joint probability that there are defects at  $\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n$  then

$$\begin{aligned} \langle JS(0) \rangle &= \sum_{\mathbf{r}_1} W(\mathbf{r}_1) [S(\mathbf{r}_1) - S(0)] \\ \langle J^2 S(0) \rangle &= \sum_{\mathbf{r}_1} \sum_{\substack{\mathbf{r}_2 \\ \mathbf{r}_1 \neq \mathbf{r}_2}} W(\mathbf{r}_1, \mathbf{r}_2) [S(\mathbf{r}_1, \mathbf{r}_2) - 2S(\mathbf{r}_1) + S(0)] \\ &\vdots \end{aligned} \quad (5.4)$$

where  $S(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n)$  is the value of the thermodynamic function for defects located at  $\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n$ .

We define  $c$ , the concentration of defects, by  $c = n/N$ . If, as  $N \rightarrow \infty$  we have

$$\langle J^k S(0) \rangle = \frac{S_k}{N^k} \quad k = 0, 1, 2, \dots \quad (5.5)$$

where the  $S_k$  are constants, and  $n \rightarrow \infty$  in such a way that  $c$  remains constant, we may write, using eq. (5.1),<sup>9)</sup>

$$\langle S(n) \rangle = S_0 + S_1 c + \frac{S_2}{2!} c^2 + \dots \quad (5.6)$$

If  $S$  is an additive function of the normal mode frequencies, as in eq. (2.1) then<sup>5)</sup>

$$\langle J^k S(0) \rangle = \frac{1}{2\pi i} \left\langle \int_C f(z) d \sum_{j=1}^K (-1)^{K-j} \binom{K}{j} \ln |J_j(z)| \right\rangle \quad (5.7)$$

where  $|J_j(z)|$  is the correction determinant associated with  $j$  defects.

The problem of averaging is a matter of some difficulty because of the interactions between the defects previously described. At sufficiently high temperatures one can ignore the correlations so that

$$W(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n) = 1 / \binom{N}{n}. \quad (5.8)$$

For example in this approximation the "interaction- $S$ " of two defects is given by

$$\langle J^2 S(0) \rangle = \frac{2}{N(N-1)} \left\langle \int_C f(z) d \ln \frac{|J_2(z)|}{|J_1(z)|^2} \right\rangle. \quad (5.9)$$

One can thus evaluate the first few terms of eq. (5.6) without difficulty.

For example if we allow the position of defect atoms to be uncorrelated, i.e., if we use eq. (5.8) to express  $W(\mathbf{r}_1)$  and  $W(\mathbf{r}_1, \mathbf{r}_2)$ , then we find for the free energy per particle,  $\langle \frac{A(T)}{N} \rangle$ , at high temperatures for a concentration  $c$  of isotope defects:

$$\begin{aligned} \langle \frac{A(T)}{N} \rangle = & \kappa T \ln \frac{\hbar \omega_L}{2\kappa T} + \kappa T \left[ \frac{1}{48} \left( \frac{\hbar \omega_L}{\kappa T} \right)^2 - \frac{1}{7680} \left( \frac{\hbar \omega_L}{\kappa T} \right)^4 + \dots \right] \\ & + c \left\{ \frac{\kappa T}{2} \ln \frac{1}{Q} + \kappa T \left[ \frac{1}{48} \left( \frac{1}{Q} - 1 \right) \left( \frac{\hbar \omega_L}{\kappa T} \right)^2 \right. \right. \\ & \left. \left. - \frac{\kappa T}{2880} \left[ \frac{1}{A} \left( \frac{1}{Q} + \frac{1}{Q^2} \right) - \frac{1}{2} \right] \left( \frac{\hbar \omega_L}{\kappa T} \right)^4 + \dots \right] \right\} \\ & + c^2 \left\{ \left[ -\frac{\epsilon^2 \kappa T}{23,040} \left( \frac{\hbar \omega_L}{\kappa T} \right)^4 + \dots \right] + O(\epsilon^3) \right\} + O(c^3) \end{aligned} \quad (5.10)$$

where we have used eqs. (3.8b) and (3.22).

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### Appendix. Change in frequency spectrum due to a single isotope defect

The mathematical methods outlined in Section II are general enough to be applicable to lattices of more than one dimension. However, in order that the  $D$ -shaped contour described in Section II can be used as the path of integration in evaluating  $\mathcal{J}S$  the function  $f(\omega)$  (cf. eq. (2.1)) must satisfy several conditions. Inasmuch as  $J(\omega)$  is a function of  $\omega^2$  only, the logarithmic derivative  $\frac{d}{d\omega} \ln J(i\omega)$  of  $J(i\omega)$  evaluated along the imaginary axis is an odd function of  $\omega$ . In order that the integration down the imaginary axis should not vanish,  $f(i\omega)$  must either be an odd function of  $\omega$  also, or it must have an odd part. Secondly, and as important, in order that the contribution to the integral from the semi-circular portion of the contour vanish in the limit as  $R \rightarrow \infty$ , it can be shown that  $f(\omega)$  must behave like at most  $|\omega|^\alpha$  as  $|\omega| \rightarrow \infty$ , where  $\alpha < 2$ . When these two conditions are not satisfied we must either re-express  $f(\omega)$  in an alternative form or modify the contour. (It is these considerations which led to the decomposition of the free energy given by eq. (2.16).) In the case of the characteristic function of the squares of the normal mode frequencies which is defined by  $f(\omega) = N^{-1} \exp(i\alpha\omega^2)$ , these two conditions are not satisfied. Choosing instead Laplace transforms for which  $f(\omega) = N^{-1} \exp(-\alpha\omega)$  leads to very difficult integrals, but appears to be the only alternative in the two and three-dimensional cases. However, for one-dimensional lattices, a special transformation of the integral representing the inverse matrix elements of Section II leads to an interesting method for evaluating changes in additive functions of normal mode frequencies in the presence of defects. We give here an outline of the method and apply it to the evaluation of the change in the frequency spectrum of a lattice containing a single isotope defect.

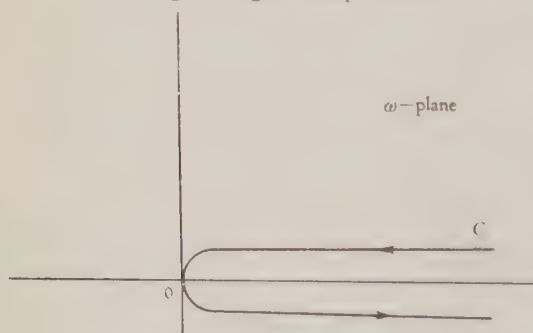


Figure A.1a. An alternative contour of integration in the  $\omega$ -plane

If we introduce the transformations\*

$$\omega = \frac{\omega_0}{2} (1 + 2 - \xi - \xi^{-1}), \quad \gamma = e^\xi, \quad (\text{A.1a,b})$$

the one-dimensional form of the expression (2.12) for the Green's function  $g(k)$  becomes

$$g(k) = \frac{1}{4\pi i} \oint_{|y|=1} \frac{y^k + y^{-k}}{(y - \xi)(y - \xi^{-1})} dy \quad (\text{A.2})$$

\* T. Tanaka, unpublished work.

$$= \frac{1}{\gamma} \frac{\xi^k}{\xi - \xi^{-1}} \quad \text{for } |\xi| < 1 \quad (\text{A} \cdot 3a)$$

$$= \frac{1}{\gamma} \frac{\xi^{-k}}{\xi^{-1} - \xi} \quad \text{for } |\xi| > 1. \quad (\text{A} \cdot 3b)$$

The transformation (A. 1a) maps the contour  $C$  in the  $\omega$ -plane (see Fig. A. 1a) into the contour  $\Gamma$  in the  $\xi$ -plane (Fig. A. 1b) where the dashed curve is the unit semicircle. The expression for  $\Delta S$  given by eq. (2.8) becomes

$$\Delta S = \frac{1}{2\pi i} \int_{\Gamma} f\left(\frac{\omega_L}{2} \sqrt{2 - \xi - \xi^{-1}}\right) d \ln \left| \mathcal{J}\left(\frac{\omega_L}{2} \sqrt{2 - \xi - \xi^{-1}}\right) \right|. \quad (\text{A} \cdot 4)$$

We now apply this formalism to the evaluation of the change in the frequency spectrum of a monatomic linear chain due to the presence of a single isotope defect. In this case  $f(\omega) = N^{-1} \exp(i\alpha\omega^2)$  and we define  $\Delta F(\alpha)$  by

$$\Delta F(\alpha) = \frac{1}{2\pi N i} \int_c e^{i\alpha\omega^2} d \ln |\mathcal{J}(\omega)|. \quad (\text{A} \cdot 5)$$

$\Delta F(\alpha)$  is related to the change in the distribution function of the squares of the normal mode frequencies,  $\Delta G(\omega^2)$ , by

$$\Delta G(\omega^2) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Delta F(\alpha) e^{-i\alpha\omega^2} d\alpha. \quad (\text{A} \cdot 6)$$

$G(\omega^2)$  is related to the usual distribution function for the normal mode frequencies,  $g(\omega)$ , by  $g(\omega) = 2\omega G(\omega^2)$ .

The determinant  $|\mathcal{J}(\omega)|$  corresponding to a monatomic linear chain with one isotope defect is obtained from eq. (3.2) as

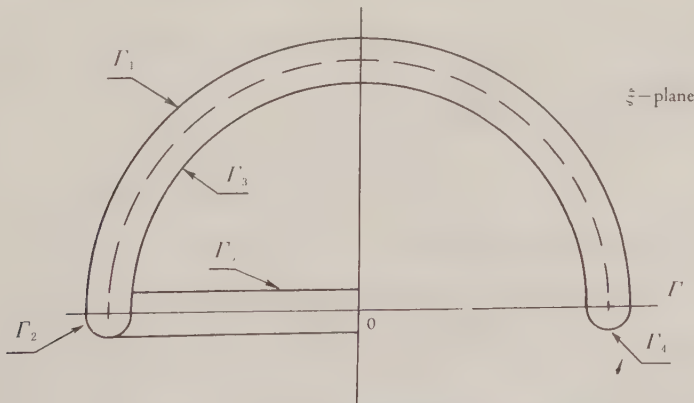


Figure A. 1b. Transformed contour of integration suitable for one-dimensional problems.

$$|\mathcal{J}(\omega)| = 1 - \varepsilon M \omega^2 g(0) \quad (\text{A} \cdot 7)$$

and in the present case becomes



$$\left| d\left(\frac{\omega_L}{2} \sqrt{2 - \hat{\xi} - \hat{\xi}^{-1}}\right) \right| = 1 - \frac{\varepsilon(2 - \hat{\xi} - \hat{\xi}^{-1})}{\hat{\xi} - \hat{\xi}^{-1}} \quad |\hat{\xi}| < 1 \quad (\text{A} \cdot 8a)$$

$$= 1 - \frac{\varepsilon(2 - \hat{\xi} - \hat{\xi}^{-1})}{\hat{\xi}^{-1} - \hat{\xi}} \quad |\hat{\xi}| > 1. \quad (\text{A} \cdot 8b)$$

To carry out the integration of (A.5) we divide up the contour  $I'$  into suitable portions as shown in Fig. A. 1b and use the appropriate value of  $\omega = \frac{\omega_L}{2} \sqrt{2 - \hat{\xi} - \hat{\xi}^{-1}}$  on each portion.

a) Integration on  $I_1$  and  $I_3$ .

On  $I_1$ ,  $\hat{\xi} = (1 + \delta)e^{i\theta}$  and  $|\hat{\xi}| > 1$ . In the limit as  $\delta \rightarrow 0$

$$(JF)_{I_1} = \frac{1}{2\pi i} \int_0^\pi e^{i\alpha\omega_L^2 \sin^2(\theta/2)} d \ln \left[ 1 + \varepsilon \frac{1 - \cos \theta}{i \sin \theta} \right]. \quad (\text{A} \cdot 9a)$$

On  $I_3$ ,  $\hat{\xi} = (1 - \delta)e^{i\theta}$  and  $|\hat{\xi}| < 1$ . In the limit as  $\delta \rightarrow 0$

$$(JF)_{I_3} = - \frac{1}{2\pi i} \int_0^\pi e^{i\alpha\omega_L^2 \sin^2(\theta/2)} d \ln \left[ 1 - \varepsilon \frac{1 - \cos \theta}{i \sin \theta} \right]. \quad (\text{A} \cdot 9b)$$

Adding these two results we obtain

$$\begin{aligned} (JF)_{I_1+I_3} &= \frac{1}{\pi} \int_0^\pi e^{i\alpha\omega_L^2 \sin^2(\theta/2)} I_m \left\{ d \ln \left[ 1 + \varepsilon \frac{1 - \cos \theta}{i \sin \theta} \right] \right\} \\ &= \frac{1}{\pi} \int_0^\pi e^{i\alpha\omega_L^2 \sin^2(\theta/2)} \frac{\cos \theta - 1}{\sin^2 \theta + \varepsilon^2 (1 - \cos \theta)^2} d\theta \\ &= - \frac{1}{2\pi} \int_0^\pi e^{i\alpha\omega_L^2 \sin^2(\theta/2)} \frac{d\theta}{(1 - (1 - \varepsilon^2) \sin^2(\theta/2))}. \end{aligned} \quad (\text{A} \cdot 10)$$

b) Integration on  $I_2$ :

We divide this into two parts,  $I_2^{(1)}$  where  $0 \leq \theta \leq (\pi/2)$  and  $I_2^{(2)}$  where  $\pi/2 \leq \theta \leq \pi$ . In either case,  $\hat{\xi} = -(1 + \delta e^{i\theta})$ .

For  $I_2^{(1)}$ ,  $|\hat{\xi}| > 1$ , and in the limit  $\delta \rightarrow 0$ ,

$$(JF)_{I_2^{(1)}} = \frac{1}{2\pi i} \int_0^{\pi/2} \exp(i\alpha\omega_L^2) (-i) d\theta = -\frac{1}{4} \exp(i\alpha\omega_L^2).$$

For  $I_2^{(2)}$ ,  $|\hat{\xi}| < 1$ , and in the limit  $\delta \rightarrow 0$

$$(JF)_{I_2^{(2)}} = \frac{1}{2\pi i} \int_{\pi/2}^\pi \exp(i\alpha\omega_L^2) (-i) d\theta = -\frac{1}{4} \exp(i\alpha\omega_L^2).$$

Hence

$$(JF)_{I_2} = -\frac{1}{2} \exp(i\alpha\omega_L^2). \quad (\text{A} \cdot 11)$$

c) Integration on  $I_4$ :

In this case, by the substitution  $\hat{\xi} = 1 - \delta e^{i\theta}$ ,  $0 \leq \theta \leq \pi$  it is easily shown that for  $\delta \rightarrow 0$ , the differential coefficient of the logarithm vanishes. The contribution from this

source therefore vanishes.

d) Integration along the rectangle  $\Gamma_5$ :

The contribution from this part of the contour is simply the sum of the residues at the poles of the integrand inside the rectangle. The poles are at the roots of the equation

$$1 - \varepsilon \frac{2 - \xi - \xi^{-1}}{\xi - \xi^{-1}} = 0. \quad (\text{A} \cdot 12)$$

There is only one root to this equation in the interval  $(-1, 0)$  and this is given by  $\xi = \frac{\varepsilon - 1}{\varepsilon + 1}$  with  $0 < \varepsilon < 1$ . If  $\varepsilon < 0$  there is no root inside the rectangle. We thus obtain

$$(\mathcal{J}F)_{\Gamma_5} = e^{i\alpha\omega_L^2/(1-\varepsilon^2)} \quad 0 < \varepsilon < 1 \quad (\text{A} \cdot 13a)$$

$$= 0 \quad \varepsilon < 0. \quad (\text{A} \cdot 13b)$$

Adding all of these results we obtain

$$\mathcal{J}F(\alpha) = - \frac{|\varepsilon|}{2\pi N} \int_0^\pi \frac{\exp(i\alpha\omega_L^2 \sin^2(\theta/2))}{1 - (1 - \varepsilon^2)\sin^2(\theta/2)} d\theta - \frac{1}{2} e^{i\alpha\omega_L^2} + e^{i\alpha\omega_L^2/(1-\varepsilon^2)} \quad 0 < \varepsilon < 1 \quad (\text{A} \cdot 14a)$$

$$= \frac{|\varepsilon|}{2\pi N} \int_0^\pi \frac{\exp(i\alpha\omega_L^2 \sin^2(\theta/2))}{1 - (1 - \varepsilon^2)\sin^2(\theta/2)} d\theta - \frac{1}{2} e^{i\alpha\omega_L^2} \quad \varepsilon < 0. \quad (\text{A} \cdot 14b)$$

If we substitute these expressions into eq. (A. 6) and interchange the order of integration we obtain

$$\begin{aligned} \Delta G(\omega^2) &= - \frac{|\varepsilon|}{2\pi N} \int_0^\pi \frac{\delta(\omega_L^2 \sin^2(\theta/2) - \omega^2)}{1 - (1 - \varepsilon^2)\sin^2(\theta/2)} d\theta \\ &\quad - \frac{1}{2N} \delta(\omega_L^2 - \omega^2) + \frac{1}{N} \delta\left(\frac{\omega_L^2}{1 - \varepsilon^2} - \omega^2\right) \quad 0 < \varepsilon < 1 \quad (\text{A} \cdot 15a) \end{aligned}$$

$$= \frac{|\varepsilon|}{2\pi N} \int_0^\pi \frac{\delta(\omega_L^2 \sin^2(\theta/2) - \omega^2)}{1 - (1 - \varepsilon^2)\sin^2(\theta/2)} d\theta - \frac{1}{2N} \delta(\omega_L^2 - \omega^2) \quad \varepsilon < 0. \quad (\text{A} \cdot 15b)$$

where  $\delta(x)$  is the Dirac delta function.

Using the following property of the delta function

$$\int_I f(x) \delta[g(x)] dx = \sum_i \frac{f(x_i)}{g'(x_i)}$$

where the summation extends over all the zeroes of  $g(x)$  in the integration interval  $I$ , we obtain finally

$$\begin{aligned} \Delta G(\omega^2) &= - \frac{|\varepsilon|}{2\pi N} \frac{\omega_L^2}{[\omega_L^2 + (\varepsilon^2 - 1)\omega^2] \omega \sqrt{\omega_L^2 - \omega^2}} \\ &\quad - \frac{1}{2N} \delta(\omega_L^2 - \omega^2) + \frac{1}{N} \delta\left(\frac{\omega_L^2}{1 - \varepsilon^2} - \omega^2\right) \quad 0 < \varepsilon < 1 \quad (\text{A} \cdot 16a) \end{aligned}$$

$$= \frac{|\xi|^2}{2\pi N} \frac{\omega_L^2}{[\omega_L^2 + (\xi^2 - 1)\omega^2]} \omega \sqrt{\omega_L^2 - \omega^2} - \frac{1}{2N} \delta(\omega_L^2 - \omega^2) \quad \xi > 0. \quad (\text{A} \cdot 16b)$$

The above expressions show that for positive  $\xi$ , i.e., a lighter isotopic defect, the inband frequencies are raised by an amount of the order of  $1/N$  and a discrete frequency appears at  $\omega = \frac{\omega_L}{\sqrt{1 - \xi^2}}$ . For a heavier defect only the inband frequencies are depressed; no discrete frequency comes out of the band.

Recently Maradudin and Weiss\* have presented a method for evaluating successive terms in the power series expansion of  $g(\omega)$  in the neighborhood of  $\omega \sim 0$ . The method is general and is not restricted to the one dimensional case. The result of their treatment is that if we can expand  $g(\omega)$  about  $\omega = 0$  as

$$g(\omega) = a_0 + a_2\omega^2 + a_4\omega^4 + \dots \quad (\text{A} \cdot 17)$$

then the coefficients  $a_{2n}$  are given by

$$a_{2n} = \frac{(-1)^n}{N\pi(2n)!} \frac{Q^{(2n)}(0)}{\omega_L^{2n+1}}, \quad (\text{A} \cdot 18)$$

where  $Q^{(2n)}(0)$  is defined by eq. (2.29).

In the particular case of a single isotope defect in a linear chain for which

$$Q(f) = \frac{-\xi}{(1+f^2)^{3/2} - \xi f(1-f^2)}, \quad (\text{A} \cdot 19)$$

eqs. (A. 18) and (A.19) give us that

$$\mathcal{J}g(\omega) = -\frac{\xi}{N\pi\omega_L} \left\{ 1 + \left( \frac{3}{2} - \xi^2 \right) \frac{\omega^2}{\omega_L^2} + 5 \left( \frac{3}{8} - \frac{\xi^2}{2} + \frac{\xi^4}{5} \right) \frac{\omega^4}{\omega_L^4} + \dots \right\}. \quad (\text{A} \cdot 20)$$

If we obtain  $\mathcal{J}g(\omega)$  from eq. (A.16) by the relation  $\mathcal{J}g(\omega) = 2\omega \mathcal{J}G(\omega^2)$  and expand this expression in powers of  $(\omega/\omega_L)$  we get the expression (A.20) exactly.

It may be mentioned here that contour integration in the  $\hat{\xi}$ -plane instead of the  $\omega$ -plane can conveniently be used for evaluation of other additive functions of normal mode frequencies for a linear chain with defects.

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## Letters to the Editor

*The opinions expressed in these columns do not necessarily reflect those of the Board of Editors. Communications should be submitted in duplicate and should be held to within 100 lines (pica type) on standard size letter paper (approx. 21×30cm.), so that each letter will be arranged into two pages when printed. Do not forget to count in enough space for formulas, figures or tables.*

### On the Consistency between the P-wave Dispersion Relation and the Experiment

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July 14, 1958

Recently Puppi et al.<sup>1)</sup> have pointed out a discrepancy between the dispersion relation for the  $\pi^-p$  scattering amplitude and the experiments. In this note we reinvestigate the dispersion relation for the  $(3, 3)$  P-wave amplitude with new data available now and show that there is a certain discrepancy similar to Puppi's case.

For this purpose we follow the analysis by Cini et al.<sup>2)</sup> in their determination of the coupling constant using the Chew-Low equation, since the P-wave dispersion relations<sup>3)</sup> are reduced to the Chew-Low equation with the point source in the static limit. Cini's analysis involves only those data at energies higher than 100 Mev (meson kinetic energy in lab. sys.) and we can now avail ourselves of some data at energies lower than that energy. According to Cini the dispersion relation for the  $(3, 3)$  amplitude is approximated to the relation

$$\left[ \frac{\sin \delta_{33} \cos \delta_{33}}{q^3} - X(\omega_q) \right] \omega_q = \frac{4}{3} f^2 + Y\omega_q, \quad (1)$$

with

$$X(\omega_q) = \frac{1}{\pi} P \int_1^\infty \frac{d\omega_p}{p^3} I_m g_3(\omega_p) \times \left[ \frac{1}{\omega_p - \omega_q} + \frac{1}{9} \frac{1}{\omega_p + \omega_q} \right], \quad (2)$$

and

$$Y = \frac{1}{\pi} \int_1^\infty \frac{d\omega_p}{\omega_p p^3 v^2(p)} I_m \left[ \frac{10}{9} g_3(\omega_p) + \frac{4}{9} g_2(\omega_p) + \frac{4}{9} g_1(\omega_p) \right] \quad (3)$$

where  $q$  and  $\omega_q$  are the momentum and the energy of the meson in the center of mass system.  $f^2$  is the rationalized re-normalized coupling constant. If we restrict ourselves below the resonance energy, eq. (1) holds with great accuracy. We can check the consistency if we substitute the empirical data for the l.h.s. of eq. (1). In calculating  $X(\omega_q)$  we use Anderson's empirical formula

$$\tan \delta_{33} = 0.2375 p^3 / (1 + 0.75 p^2) (1.95 - \omega_p) \quad (4)$$

for the energy less than 300 Mev. The numerical calculations are done with great care, because there is a principal integral in  $X(\omega_q)$ . The results are shown in Figs. 1 and 2. In Figs. 3 and 4 we plot the

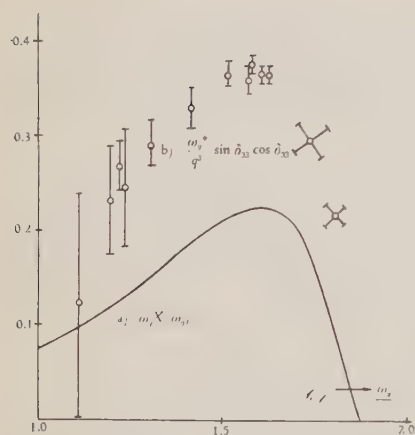


Fig. 1. a)  $\omega_\eta X(\omega_\eta)$  as obtained from numerical calculation. b) The experimental values<sup>4)</sup> of the quantity  $\omega_\eta (\sin \delta_{33} \cos \delta_{33})$ .

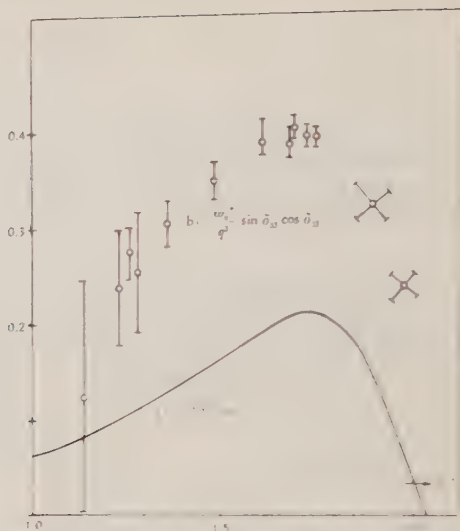


Fig. 3. a)  $\omega_\eta^* X(\omega_\eta^*)$  as obtained from numerical calculation. b) The experimental values<sup>4)</sup> of the quantity  $\omega_\eta^* (\sin \delta_{33} \cos \delta_{33})$ .

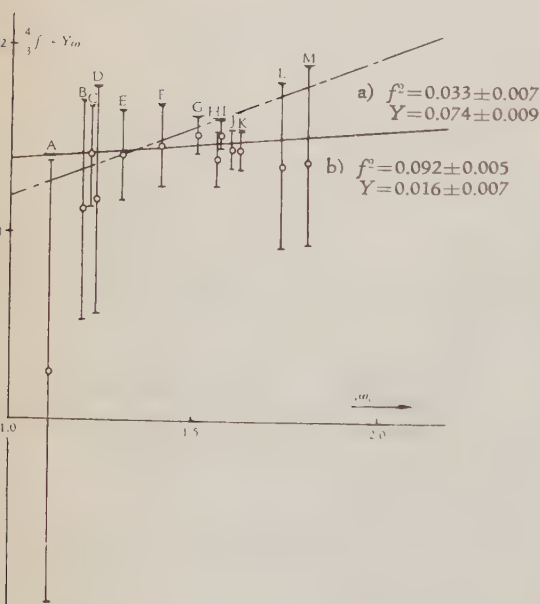


Fig. 2. The difference of curve a) and points b) in Fig. 1 plotted versus  $\omega_\eta$ .

- a)  $f^{\pi 2} = 0.033 \pm 0.007$ ,  $Y = 0.074 \pm 0.009$ , evaluated with a weighted least square fit to the points below 100 Mev (A~G).  
 b)  $f^{\pi 2} = 0.092 \pm 0.005$ ,  $Y = 0.016 \pm 0.007$ , evaluated with a weighted least square fit to the points below resonance (A~M).

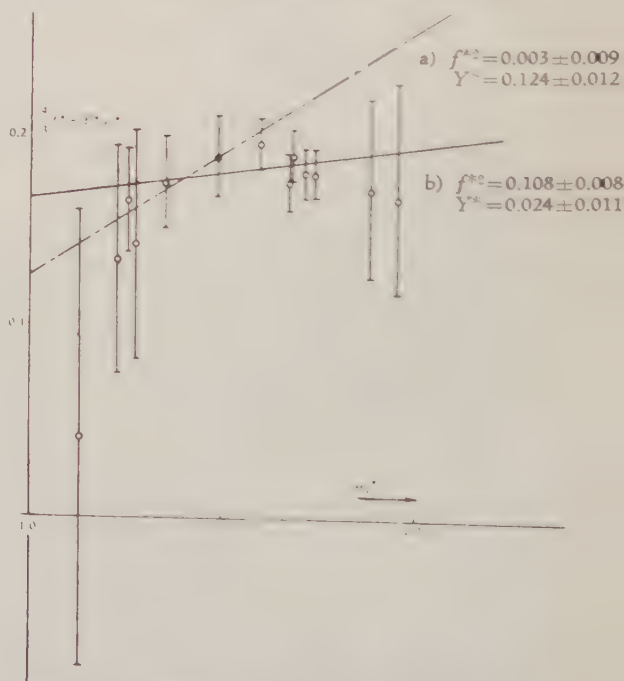


Fig. 4. The difference of curve a) and points b) in Fig. 3 plotted versus  $\omega_\eta^*$ .

- a)  $f^{\pi 2} = 0.108 \pm 0.008$ ,  $Y^{\pi 2} = 0.024 \pm 0.011$ , evaluated with weighted least square fit to the points below 100 Mev (A~G).  
 b)  $f^{\pi 2} = 0.003 \pm 0.009$ ,  $Y^{\pi 2} = 0.124 \pm 0.012$ , evaluated with a weighted least square fit to the points below resonance. (A~M).



results obtained by replacing  $\omega$  by  $\omega^* = \omega + k^2/2M$ , as is usually done in the analysis of the *Chew-Low* equation.

As is seen in the figures, if we use all the data below the resonance ( $A \sim M$ ), we obtain

$$f^2 = 0.092 \pm 0.005$$

$$\text{with } Y = 0.016 \pm 0.007,$$

$$\text{and } f^{*2} = 0.108 \pm 0.008$$

$$\text{with } Y^* = 0.024 \pm 0.011,$$

where the symbol \* means the replacement  $\omega \rightarrow \omega^*$ . The values are consistent with the one authorized today. If, however, we restrict us in the energy range less than 100 *Mev*, we obtain

$$f^2 = 0.033 \pm 0.007$$

$$\text{with } Y = 0.074 \pm 0.009,$$

$$\text{and } f^{*2} = 0.003 \pm 0.009$$

$$\text{with } Y^* = 0.124 \pm 0.012$$

These values are far less than 0.08. The above constants are determined by the method of weighted least squares. The errors are the standard mean square deviations, which do not involve explicitly the experimental errors in itself. Thus, we can conclude that for the *P-wave* amplitudes there also seems to be a discrepancy similar to the one pointed out by Puppi, so long as we use Anderson's empirical formula in calculating  $X(\omega_\eta)$ . On the other hand, there are many facts indicated that the theory would reproduce the empirical facts quantitatively so far as only *P-waves* are concerned with low energy. Then we believe that this discrepancy would not lead to the immediate break down of the micro causality, since the integral  $X(\omega_\eta)$  seems to be sensitive to the derivative of the curve of  $\delta_{\eta\eta}$  and the discrepancy may

vanish if we use the modified formula, increasing more rapidly than Anderson's at 100 *Mev*, which is the same situation as is indicated by Lommon et al.<sup>(5)</sup> on Puppi's case. We wish to thank Professor H. Tanaka for valuable discussions.

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### On the Radiative Corrections to the Scattering of Mesons in an External Electromagnetic Field\*

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The radiative corrections to the scattering of a spin-zero particle in an external electromagnetic field were considered some time ago by Kinoshita and Nambu<sup>1)</sup>. These authors used the Duffin-Kemmer formalism,<sup>2)</sup> thereby also obtaining the corresponding result for spin-one particles. In view of the complexity of their calculations it was felt desirable to recalculate the effects in question for the more important case of spin-zero particles, by a different method.

Such a method is available in the Klein-Gordon formalism.<sup>3)</sup> On computation one finds that the second order radiative corrections lead, in first Born approximation in the external field, to a modification  $j_\mu'(x) = j_\mu(x) + \delta j_\mu(x)$  of the free meson current  $j_\mu(x)$ . Here

$$\begin{aligned} \delta j_\mu(x) = & \frac{\alpha}{4\pi} \int d^4x' j_\mu(x') \left[ \left( -2 + \frac{\square^2}{m^2} \right) \right. \\ & \times E(x-x') + \left( \log \frac{m}{2k_{\min}} \right) \frac{\square^2}{m^2} \{ F_0(x-x') \\ & \left. + F_1(x-x') \} - \frac{1}{6} \frac{\square^2}{m^2} F_2(x-x') \right] \quad (1) \end{aligned}$$

where the notation is as in reference 1.

Equation (1) differs from the corresponding result of Kinoshita and Nambu (see ref. 1, eq. (140) and eq. (65)) by a factor of 2 in the term proportional to  $E(x-x')$ . This discrepancy may be traced (with some labor) to an error<sup>4)</sup> in eq. (133) of ref. 1. When this is rectified the calculations are in complete agreement, as is to be expected on theoretical grounds<sup>5)</sup>.

One of us (A.K.B.) would like to thank Professor R. C. Majumdar for suggesting this problem and for helpful discussions; he is grateful to the University of Delhi, where this work was begun, for a research assistantship. We appreciate the interest

that Professor T. Kinoshita has shown in this work.

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- 4) On computation of eq. (133) from eq. (132) of Ref. 1, one finds that the term written as

$$\left( 4 + \frac{2p^2}{m^2} \right) \frac{\log \left( 1 + \frac{p^2}{4m^2} (1-r^2) \right)}{1 + \frac{p^2}{4m^2} (1-r^2)}$$

should be divided by 2. The results given in Ref. 1, eq. (136), for the vector case should also be changed accordingly.

- 5) K. V. Roberts, Phys. Rev. 83 (1951), 188.

## On Parity Nonconservation

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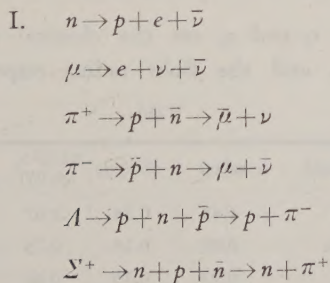
June 6, 1958

Parity nonconservation is established in various decay processes. Questions arise; why do there exist parity conserving interactions and nonconserving ones, what does parity nonconservation mean and is it a characteristic to the weak interaction as was expected? I imagine that parity nonconservation has its origin in the stereographical structure of the form factor in the primary interaction in which four particles participate, in a circumstance similar to that in the

structure of  $d$  and  $l$  isomers in stereochemistry. I have not yet reached to a concrete conclusion, but the following fact seems to me to suggest the idea and to have a phenomenological meaning by itself.

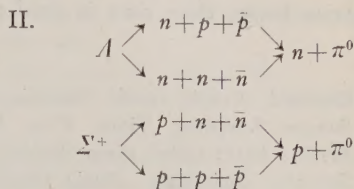
The experimental results, (a) the asymmetry parameter ( $\alpha$ ) is large in  $\beta$  decay,  $\mu$  decay,  $\pi$  decay and  $\Lambda \rightarrow p + \pi^-$  decay and (b) parity is not conserved in  $\Sigma^+$  decay while the up-down asymmetry is very small in  $\Sigma^-$  decay<sup>1)</sup> (although the observed values of asymmetry coefficient ( $P\alpha$ ) are not yet accurate), can be explained if we assume that (1) the interaction of four Fermi particles (including Hyperons ( $Y$ )) is the only primary weak interaction, (2) parity is not in general conserved in the transition by this interaction but parity is conserved in the exceptional case in which there are indistinguishable particles—particles to be excluded by the Pauli principle—in the particles outgoing from or ingoing to a (nonlocal) vertex of this interaction. (It is the case when  $a$  and  $c$  are identical particles in  $b \rightarrow a + c + \bar{d}$ .) and (3) the interaction ( $N + \bar{N} \rightarrow \pi$ ) is considerably stronger than the interaction ( $Y + \bar{Y} \rightarrow \pi$ ). (The assumption (3) makes the discussion very simple and definite.)

These assumptions give the following decay processes.

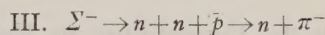


In these cases, no indistinguishable particles are present in every stage connected by the

weak interaction, hence the asymmetry parameter is large for these cases.



In these case, both the parity conserving process (lower process) and nonconserving process (upper process) are possible. For these cases, the asymmetry parameter is expected to be relatively small compared with those of I.



In this case, there is only a parity conserving process, hence the asymmetry parameter is expected to be very small.

In view of field theory, it may be difficult to construct the desired form factor which gives the interaction considered, since the interaction Lagrangian must receive certain severe restriction originated from the assumed frame of space-time. In this connection, a new description of the dynamics of elementary particles might become necessary. In field theory, the system of elementary particles are represented by a system of some operator functions of the parameter  $x$  and the dynamics is the prescription of the variation with respect to the parameter  $x$ , of the state vector or the operator functions. (In this sense, the parameter  $x$  has the meaning of space-time.) I think it is reasonable to formulate the dynamics of elementary particles without referring to the parameter  $x$ .<sup>2)</sup> In this case,  $x$  is a kinematical parameter (relating to the momentum of particles) and has not directly the meaning of space-time (Contrary to field theory, space-time is derived from



the dynamics as a statistical quantity), and the restriction for the interaction operator becomes looser than that in field theory.

- 1) Crawford, Cresti, Good, Gottstein, Lyman, Solmitz, Stevenson, Ticho, *Phys. Rev.* **108** (1957), 1102; Eisler, Plano, Prodel, Samios, Schwartz, Steinberger, Bassi, Borelli, Puppi, Tanaka, Woloshek, Zoboli, Conversi, Franzini, Mannelli, Santangelo, Silvestrini, Glaser, Graves, Perl, *Phys. Rev.* **108**, (1957), 1353.
- 2) T. Tati, *Nuovo Cimento* **4** (1956), 75, *Prog. Theor. Phys.* **18** (1957), 235.

## On the Positronium in Metals

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The experimental results indicate that in metals positrons reach the thermal velocity in a short time and subsequently remain free, annihilating with conduction electrons.<sup>1)</sup> However, the possibility of the existence of positronium in metals, undergoing continuous exchange of the electron partner may not be excluded. Though this problem is a many-body problem similar to that of the exciton, we consider a system of an electron and a positron and investigate the properties of the positronium in metals. The properties of the positronium in a metal are somewhat different from those of the positronium in free space. The difference is due to that the interaction between an electron and a positron in a metal is

not the Coulomb interaction, but a screened short ranged interaction.<sup>2)</sup>

Choosing the Sommerfeld free electron theory as a starting basis, we have, for the non-relativistic Schrödinger equation for the positronium in a metal

$$\left(\frac{p^2}{\mu} - e^2 \frac{e^{-qr}}{r}\right) \psi = E \psi, \quad (1)$$

where  $\mu = 2m_p m_e / (m_p + m_e)$ ,  $m_p$  and  $m_e$  being the masses of the positron and the electron respectively. For the ground state (S state) (1) reduces to

$$\frac{d^2 R}{d\tilde{z}^2} + b \frac{e^{-\tilde{z}}}{\tilde{z}} R + \gamma R = 0, \quad (2)$$

where  $R(r) = \psi(r)/r$ ,  $\tilde{z} = qr$ ,  $b = e^2 \mu / b^2 q$  and  $\gamma = E \mu / b^2 q^2$ . The screened potential  $e^{-\tilde{z}}/\tilde{z}$  can conveniently be approximated by Hulthén potential  $e^{-\tilde{z}}/(1-e)$ . Then the normalized eigenfunction is given by<sup>3)</sup>

$$\begin{aligned} \psi(r) &= \{b(b^2 - 1)q/8\pi\}^{1/2} \\ &\times e^{-(b-1)qr/2} (1 - e^{-qr})/r, \end{aligned} \quad (3)$$

and the eigenvalue is

$$E = -b^2 q^2 / 4\mu \cdot (1 - e^2 \mu / b^2 q)^2. \quad (4)$$

The lifetime for the two-photon decay is given by<sup>4)</sup>

$$\begin{aligned} \tau &= 1/\pi c r_0^2 |\psi(0)|^2 \\ &= 1.25 \times 10^{-10} / 8\pi a_0^3 |\psi(0)|^2 \text{ sec.} \end{aligned} \quad (5)$$

where  $r_0$  and  $a_0$  are the classical electron radius and the Bohr radius respectively.

Table

Metal	$m^*/m$	$-E$ (ry)	$8\pi a_0^3 \times  \psi(0) ^2$	$\lambda/2a_0$
Li	1.45	0.34	2.67	0.72
Na	0.98	0.14	0.75	1.05
K	0.93	0.15	0.66	1.10
Rb	0.89	0.14	0.57	1.16
C	0.83	0.16	0.44	1.15

In the Table we have computed the binding energy and the lifetime, assuming  $m_p = m_e = m^*$ . The binding energies of positroniums in alkali metals are smaller than that of the positronium in free space (0.5 ry), and the lifetimes are longer except in Li.

$\lambda$  is defined by  $\phi(\lambda)/\phi(0) = e^{-1}$ .  $q$  is determined by the relation<sup>5)</sup>  $q/k_0 = 0.470 r_s^{1/2}$ , where  $k_0$  is the radius of the Fermi sphere and  $r_s = (3/4\pi n a_0^3)^{1/3}$ ,  $n$  and  $a_0$  being the electron density and the Bohr

radius respectively.

- 1) S. Berko and F. I. Hereford, *Rev. Mod. Phys.* **28** (1956), 299.  
R. A. Ferrell, *ibid.* **28** (1956), 308.
- 2) D. Bohm and D. Pines, *Phys. rev.* **92** (1953), 609.
- 3) L. Rosenfeld, *Nuclear Forces* (North-Holland Publishing Company, 1948), 77.
- 4) J. M. Jauch and F. Rohrlich, *The Theory of Photons and Electrons* (Addison-Wesley Publishing Company, 1955), 286.
- 5) K. Sawada et al, *Phys. Rev.* **108** (1957), 507.



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